1-1-1986

Angular-Distribution Of Fluorescence From Photoionization-Produced He+ (N=2)

J. Jiménez-Mier

C. Denise Caldwell
University of Central Florida

D. L. Ederer

Angular Distribution of Fluorescence from Photoionization-Produced He^+ $(n=2)$

J. Jiménez-Mier[(a)]
Physics Department, Yale University, New Haven, Connecticut 06511

C. Denise Caldwell
Department of Physics, University of Central Florida, Orlando, Florida 32816

and

D. L. Ederer
Radiation Physics Division, National Bureau of Standards, Gaithersburg, Maryland 20899

(Received 30 June 1986)

We report the first measurement of the angular distribution of the 304-Å $He^+(n=2)$ radiation following photoionization. This distribution reflects the alignment of the ion, which is related to the fraction $\xi=\sigma(2p,kd)/[\sigma(2p,ks)+\sigma(2p,kd)]$ of d component in the electron wave. The experimental angular distributions correspond to alignments of -0.62 ± 0.03 and -0.62 ± 0.02 at photon energies of 65.5 and 66.5 eV, respectively. These translate into ratios $\xi=0.25\pm0.04$ and 0.25 ± 0.03, in good agreement with close-coupling calculations.

PACS numbers: 32.80.Fb

The helium atom is to the two-electron problem what the hydrogen atom is to the one-electron problem. Because of the simplicity of the interaction term, effects of electron-electron correlation are much more readily available theoretically. Experimentally, the ease of handling helium, together with the development of new techniques, has led to numerous and better results for parameters associated with photoionization, where correlation effects are highly pronounced.[1] Experimental results are available for the total photoionization cross section,[2] the partial photoionization cross section leaving the ion in the $n=2$ state,[3-6] the $n=2$ subshell branching ratio $R=\sigma(2p)/\sigma(2s)$,[7] and the $n=2$ photoelectron asymmetry parameter[8-11] β. Of these quantities R and β are the most sensitive to finer details of the electron-electron correlation.

To date no experimental determination is available for the complementary parameter to β, namely, the alignment A_0 of the resulting He^+ ion.[12] According to Fano and Macek,[13] the alignment may be parametrized in terms of the distribution of angular momentum j as

$$A_0(j)=\frac{\sum_{m_j}\sigma(j,m_j)[3m_j^2-j(j+1)]}{j(j+1)\sum_{m_j}\sigma(j,m_j)}, \qquad (1)$$

where j is the total angular momentum of the excited state of the ion and $\sigma(m_j)$ is the partial cross section for production of the m_j sublevel of that state. In the helium photoionization channel leading to production of the ion in the $2p\,{}^2P_{3/2}$ level the outgoing electron wave will consist of both s and d components. The alignment will be determined by the relative proportion of each of these components. The fractional amount of d wave can be expressed in terms of the partial cross section for each component by[14] $\xi=\sigma(2p,kd)/[\sigma(2p,ks)+\sigma(2p,kd)]$. As LS coupling may be presumed to be valid, the connection between A_0 and ξ follows from Eq. (1) by application of the transformation

$$\sigma(j,m_j)=\textstyle\sum_{m_l}|\langle l,m_l;s,m_j-m_l\,|\,jm_j\rangle|^2\sigma(l,m_l).$$

The symbol $\langle l,m_l;s,m_j-m_l\,|\,jm_j\rangle$ is a Clebsch-Gordan coefficient, and $\sigma(l,m_l)$ is the partial cross section for production of the m_l sublevel of the orbital angular-momentum state l. The result for $l=1$ is[15]

$$A_0(\tfrac{3}{2})=-\tfrac{4}{5}+\tfrac{18}{25}\xi,\;\; A_0(\tfrac{1}{2})=0. \qquad (2)$$

For photoionization leading to production of the $n=2$ final state three open channels exist. In the angular-momentum representation we write these as $(2s,kp)$, $(2p,ks)$, and $(2p,kd)$, where the first entry refers to properties of the bound electron and the second to those of the continuum electron. Within this basis there are five real parameters making up the density matrix for the system, the three diagonal elements plus two relative phases δ_{12} and δ_{23} between the channels. From the $n=2$ cross section and the ratio R we obtain two relationships among the diagonal elements. From the alignment we obtain ξ, the third quantity necessary to evaluate all three diagonal elements independently.

Ideally, we would like to extract information about two diagonal elements and one phase from a measurement of β for the $2p$ state. However, this is difficult in the case of helium, as the interpretation of β from experiment is closely linked to the value of R. Because of the small energy splitting between the $2p$ and $2s$ states (14 GHz), photoelectrons which leave the ion in the $2p$ state cannot be resolved from those which leave the ion in the $2s$ state. Thus the total β is a sum of two contributions, β_{2s} corresponding to production of the $2s$ state, and β_{2p} corresponding to the production of the $2p$ state of He^+, i.e.,

$$\beta = \frac{\sigma(2s)\beta_{2s} + \sigma(2p)\beta_{2p}}{\sigma(2s)+\sigma(2p)}.$$

From symmetry arguments it is known that $\beta_{2s}=2$; however, because of the small energy splitting, a result for β_{2p} can only be obtained by folding into the analysis a theoretical or experimental result for R. Nevertheless, β_{2p} has been measured[8-11] as a function of energy above the $n=2$ threshold, and a combination of β_{2p} with the results for ξ can be used to obtain a value of the phase difference δ_{23}.

For the case of production of $He^+(n=2)$ the alignment is much easier to interpret from experiment than is β. This comes about because the alignment is a property of the $2p$ state alone, and the influence of the $2s$ state can be removed experimentally. The $2s$ state is metastable to photon decay ($\tau=1.9$ msec), and fluorescence from the $2p$ state ($\tau=0.1$ nsec) can be isolated from eventual $2s$ quenching. This fact was utilized by Woodruff and Samson[7] for measurement of the ratio R. As alignment information for helium is obtained from the fluorescence emitted by the excited-state ion as well, the influence of the $2s$ state can be subtracted.

Generally, alignment measurements have been carried out by determination of the degree of linear polarization of the fluorescence. However, since the fluorescence from the $2p$ state of He^+ lies in a region of the spectrum where an efficient measurement of the polarization is extremely difficult, we determine the alignment from the angular distribution of the fluorescence. The radiation which we observe is that at 304 Å from the process

$$\begin{aligned} &He(1s^2\,{}^1S_0) + h\nu(E > 65.4\text{ eV}) \rightarrow He^+(n=2) + e^-, \\ &He^+(2p\,{}^2P_{1/2,3/2}) \rightarrow He^+(1s\,{}^2S_{1/2}) + h\nu'(304\text{ Å}). \end{aligned} \tag{3}$$

The anisotropy in the fluorescence is parametrized in terms of the alignment A_0 according to

$$\frac{dI}{d\Omega} = \tfrac{1}{3} I_{0j}\left\{1 + \frac{1}{8}\frac{j+1}{2j-1}A_0(j)(1+3P_s\cos 2\theta)\right\}, \tag{4}$$

where I_{0j} is the total fluorescence from the state j and P_s is the linear polarization of the ionizing radiation.

For these first measurements we chose two photon energies near threshold, 65.5 and 66.5 eV. We did this because for some time a controversy existed between the results of close-coupling calculations[16] and many-body calculations[17] for the ratio R at threshold. At the time of this writing, this difference has been largely resolved in favor of the close-coupling results by the most recent measurements[7-11] of R and β.

The experiment was carried out with the high-throughput toroidal grating monochromator[18] in beam line 1 of SURF-II at the National Bureau of Standards. Light from the monochromator passes through a thin-film aluminum filter, then, after emerging from the exit slit, falls onto a pseudobeam of helium effusing from a capillary. Gas pressure is measured with an ionization gauge, and the reading is corrected for the low efficiency of response for helium. The 304-Å fluorescence is detected at right angles to the ionizing beam with a proportional counter[19] separated from the main chamber by a second Al filter 1000 Å thick. The angular distribution is measured by rotation of the interaction chamber about an axis lying along the direction of incidence of the synchrotron beam. The entire chamber rotates on a specially constructed UHV rotary flange which is a modification of a design by Silverman.[20] Vacuum changes during rotation are of the order of 4×10^{-9} Torr, and the angular position of the detector is reproducible within 1°. The quantization axis is along the direction of linear polarization of the synchrotron light.

The angular-distribution measurements reported here were carried out with a helium pressure of 2×10^{-4} Torr and 60 Torr of methane in the proportional counter. Typical counting rates were on the order of 10–20 counts/sec, with a signal-to-background ratio of 1:1. The polarization of the synchrotron radiation was measured with a single–gold-mirror reflecting polarimeter[21] installed in the experimental chamber after the helium measurements were finished.

For our experiment the equation for the angular distribution of the fluorescence follows from Eq. (4) and takes the form

$$dI/d\Omega = \tfrac{1}{3}I_0(2p)[1+\tfrac{5}{48}A_0(\tfrac{3}{2})]\{1+(15A_0(\tfrac{3}{2})P_s/[5A_0(\tfrac{3}{2})+48])\cos 2\theta\} + I_0(2s), \tag{5}$$

where θ is the angle that the detector makes with the quantization axis, and it is assumed that the population of the $j=\frac{3}{2}$ fine-structure level is twice that of the $j=\frac{1}{2}$ fine-structure level. The term $I_0(2s)$ comes from the fraction of the ions left in the $2s$ state that are quenched by collisions with neutral helium atoms.[22]

For the two excitation energies, 65.5 and 66.5 eV, nine sets of measurements were taken, each for thirteen angles between −90° and 90°. Five separate runs were used to calibrate the background as a function of storage-ring beam current, ionizing radiation energy, and detector angle. A measurement below threshold was then subtracted from the total signal at each energy point in order to get a true fluorescence intensity. The data were further normalized with respect to the storage-ring beam current. A function of the form $\kappa(1+\alpha\cos 2\theta)$ was fitted to the experimental points, with κ and α as parameters. To subtract $I_0(2s)$ from A_0, the fluorescence at 0° with and without an applied electric field was measured, and from the known branching ratio[6,11] the fraction of $2s$ ions quenched was obtained. It was found that 5% of the total

TABLE I. Results for the alignment and partial-wave ratio ξ. The normalized $|m_j| = \frac{1}{2}, \frac{3}{2}$ and $|m_{li}| = 0,1$ sublevel populations are also shown.

Energy (eV)	A_0 Meas.	A_0 Calc.[a]	ξ Meas.	ξ Calc.[b]	$^2P_{3/2}$ populations $m_j = \pm\frac{3}{2}$	$^2P_{3/2}$ populations $m_j = \pm\frac{1}{2}$	m_{li} populations $m_{li} = \pm 1$	m_{li} populations $m_{li} = 0$
65.5	-0.62 ± 0.03	-0.61	0.25 ± 0.04	0.26	0.056 ± 0.008	0.494 ± 0.008	0.075 ± 0.012	0.85 ± 0.02
66.5	-0.62 ± 0.02	-0.59	0.25 ± 0.03	0.29	0.057 ± 0.007	0.443 ± 0.007	0.075 ± 0.007	0.85 ± 0.02

[a]This number was calculated with the results from Ref. 24 using Eq. (3).
[b]Reference 24.

signal without electric field came from the collisional quenching of the $2s$ ions.

From the measured angular distributions and the fit parameters α and κ we obtain the alignment at the two energies of interest. Our results are given in Table I. The experimental angular distribution and theoretical fit for the photon energy of 66.5 eV are shown in Fig. 1. The errors quoted reflect both statistical fluctuations and systematic errors in the background subtraction. Also shown in Table I are the values of ξ obtained from A_0 via Eq. (2).

By combining the results for ξ with those for R, which we obtain by extrapolating the experimental and theoretical curves[7] to threshold, we calculate the normalized diagonal elements of the density matrix. In addition, from the values of ξ, together with reported results of the $2p$ asymmetry parameter β_{2p},[10] we can determine the relative phase δ_{23}.[14] The results for the diagonal elements of the density matrix and the phase δ_{23} are shown in Table II. As these parameters are determined by extrapolation of the experimental results, we do not quote errors, as we do not know the error to associate with the extrapolation.

The alignment given in Table I reflects the population distribution of the $j = \frac{3}{2}$, m_j sublevels of the ionic state, which, for helium, are equivalent to the partial cross sections for production of the m_j state. [See Eq. (1).] As indicated earlier, the partial cross sections for the production of the m_j sublevels are related to those for production of the $m_{li} = 0$ and $m_{li} = \pm 1$ sublevels of the state of total orbital angular momentum $l_i = 1$ of the ion. For ionization with linearly polarized radiation, the projections of the states of angular momentum of the ion are opposite to those for the photoelectron; i.e., $\sigma(m_{li} = 0) = \sigma(m_{le} = 0)$ and $\sigma(m_{li} = \pm 1) = \sigma(m_{le} = \mp 1)$. We have calculated these distributions for the ionic state and included them in Table I as well.

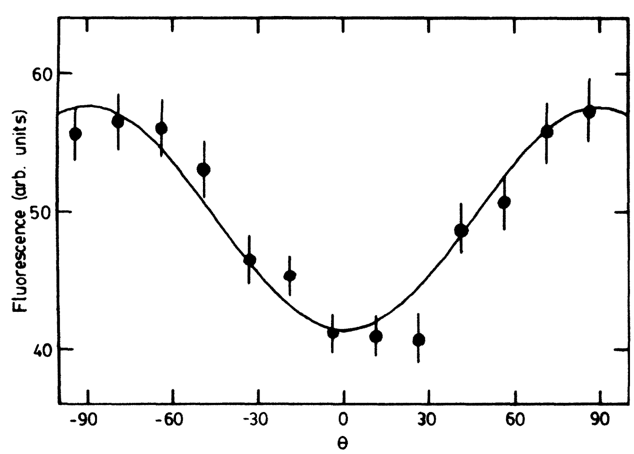

FIG. 1. Angular distribution of fluorescence following photoionization at 66.5 eV. Circles represent the experimental points with associated errors, and the solid line represents the fit $\kappa(1 + \alpha \cos 2\theta)$. At this energy the curve is given by $I(\theta) = 49.5(1 - 0.162 \cos 2\theta)$, for $P_s = 0.86$.

We see that production of ionic states with orbital angular momentum $m_{li} = 0$ is preferred. There are two contributions to the populations of the $m_{li} = 0$ sublevel. The dominant contribution comes from the ejection of s-wave photoelectrons while the other comes from the ejection of photoelectrons in d waves with projection $m_{le} = 0$. Population of the $m_{li} = \pm 1$ states only arises as photoelectrons are ejected in a d wave. Production of just s-wave photoelectrons should lead to a maximal alignment of the system; i.e., $A_0 = -0.80$. The d-wave contribution reduces this value, and our result of $A_0 = -0.62$ indicates that the amount of d wave in the outgoing electron wave is considerable.

An analysis of the dipole coupling term[23] of the electron-electron interaction shows a large mixing of the angular-momentum channels at the energies we have chosen. Ojha[24] has calculated the contribution of the d wave using a close-coupling quantum-defect analysis. He finds a total component of 26%. This result is in excellent agreement with the experimental data, providing another proof of the validity of the close-coupling calculations for this process.

We have demonstrated that the alignment measurement is an excellent probe of the helium photoionization

TABLE II. Parameters of the $n = 2$ density matrix.

Energy (eV)	$\sigma(2s,kp)$[a]	$\sigma(2p,ks)$[a]	$\sigma(2p,kd)$[a]	δ_{23}[b]
65.5	0.24	0.61	0.15	0.14
66.5	0.25	0.60	0.15	0.52

[a]By combination with the values of R obtained by extrapolation of the experimental results of Ref. 7.
[b]By combination with the values of β_{2p} obtained by extrapolation of the experimental results of Ref. 10.

leading to $He^+(n=2)$. We have reported here results for only two energies, while information for other parameters associated with the photoionization process is available throughout the energy region from the $n=2$ to the $n=3$ ionization limits. Of particular important as regards correlation should be the $3s3p$ resonance at 69.9 eV. The continued analysis of the alignment at all energies up to the $n=3$ limit is currently being planned. More importantly, however, as the $2s$ state can be separated from the $2p$ state, an alignment measurement in the presence of an external electric field may provide a means by which the second relative phase δ_{12} can be experimentally determined.

Two of us (J.J.M. and C.D.C.) thank the staff at the SURF-II facility for their hospitality and for their invaluable support, especially R. P. Madden for many stimulating discussions and R. Stockbauer for the use of and assistance with the monochromator. This research is supported by the National Science Foundation under Grants No. PHY-8307158 and No. PHY-8518598. One of us (J.J.M.) was also supported in part by Centro de Estudios Nucleares, Universidad Nacional Autónoma de México.

[a]Permanent address: Centro de Estudios Nucleares, Universidad Nacional Autónoma de México, México 04510, D.F., México.

[1]R. P. Madden and K. Codling, Astrophys. J. **141**, 364 (1965).

[2]J. B. West and G. V. Marr, Proc. Roy. Soc. London, Ser. A **349**, 397 (1976).

[3]J. A. R. Samson, Phys. Rev. Lett. **22**, 693 (1969).

[4]M. O. Krause and F. Wuilleumier, J. Phys. B **5**, L143 (1972).

[5]P. R. Woodruff and J. A. R. Samson, Phys. Rev. Lett. **45**, 110 (1980).

[6]F. Wuilleumier, M. Y. Adam, N. Sandner, and V. Schmidt, J. Phys. (Paris), Lett. **41**, 373 (1980).

[7]P. R. Woodruff and J. A. R. Samson, Phys. Rev. A **25**, 848 (1982).

[8]J. M. Bizau, F. Wuilleumier, P. Dhez, D. L. Ederer, T. N. Chang, S. Krummacher, and V. Schmidt, Phys. Rev. Lett. **48**, 588 (1982).

[9]V. Schmidt, H. Derenbach, and R. Malutzki, J. Phys. B **15**, L523 (1982).

[10]P. Morin, M. Y. Adam, I. Nenner, J. Delwiche, M. J. Hubin-Franskin, and P. Lablanquie, Nucl. Instrum. Methods Phys. Rev. **208**, 761 (1983).

[11]D. W. Lindle, T. A. Ferrett, U. Becker, P. H. Kobrin, C. M. Truesdale, H. G. Kerkhoff, and D. A. Shirley, Phys. Rev. A **31**, 714 (1985).

[12]C. D. Caldwell and R. N. Zare, Phys. Rev. A **9**, 2383 (1977).

[13]U. Fano and J. Macek, Rev. Mod. Phys. **45**, 553 (1973).

[14]P. Scott and P. G. Burke, J. Phys. B **17**, 1321 (1984).

[15]J. Jiménez-Mier, Ph.D. thesis, Yale University, 1986 (unpublished).

[16]K. A. Berrington, P. G. Burke, W. C. Fon, and K. T. Taylor, J. Phys. B **15**, L603 (1980).

[17]T. N. Chang, J. Phys. B **13**, L551 (1980).

[18]R. Stockbauer and R. P. Madden, Nucl. Instrum. Methods Phys. Rev. **195**, 207 (1982).

[19]Gas Flow Proportional Counter, J. E. Manson Co., P.O. Box 1288, Concord, MA 01742.

[20]P. J. Silverman, J. Vac. Sci. Technol. A **2**, 76 (1984).

[21]K. Rabinovitch, L. R. Canfield, and R. P. Madden, Appl. Opt. **4**, 1005 (1965).

[22]M. H. Prior and E. C. Wang, Phys. Rev. A **9**, 2383 (1974).

[23]M. J. Seaton, Proc. Phys. Soc. (London) **77**, 174 (1961).

[24]P. C. Ojha, J. Phys. B **17**, 1807 (1984).