Near-threshold measurements of K^- two-electron photoionization cross sections

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The absolute cross sections for two-electron photoionization have been measured for K^- from 70 meV below to 250 meV above the threshold by measuring product K^+ ions. To minimize complications from the photodetachment and field ionization, the electric field strength used to separate K^+ ions was limited to 300 V/cm, which ionizes only Rydberg neutral atoms with $n \geq 30$. The data fit to $\sigma \sim E^m$ (E is the total energy of the two electrons) yielded $m = 1.16 \pm 0.05$ with a reduced $\chi^2 = 0.69$, in agreement with the Wannier theory $m = 1.127$ [G. H. Wannier, Phys. Rev. **90**, 817 (1953)]. However, a nearly equally satisfactory fit can also be made to the oppositional theory of A. Temkin [Phys. Rev. A **30**, 2737 (1984)].

I. INTRODUCTION

The threshold behavior of cross sections for two slow electrons escaping from a positively charged core has been the subject of various experimental and theoretical studies. Wannier[1] first showed classically that this threshold behavior should follow an E^m law (E is the total energy of the two electrons) with $m = 1.127$ for a singly charged core. The theory was based on the physical picture in which, just above the threshold, both electrons escape in nearly opposite directions while maintaining equal distances from the core. Rau[2] and Peterkop[3] confirmed Wannier's results quantum mechanically by using asymptotic wave functions in hyperspherical coordinates expanded about the "Wannier ridge." These early theoretical treatments considered only systems with $^1S^e$ states. Using similar techniques, Klar and Schlecht,[4] Greene and Rau,[5] and Feagin,[6] extended the theory to arbitrary angular momenta, spins, and parities.

On the other hand, Temkin[7,8] employed a complementary physical picture (Coulomb-dipole) in which one electron is assumed to be generally closer to the core than the other as they both escape. Consequently, the inner electron experiences a Coulomb field, whereas the outer electron experiences an oscillating, or time-dependent dipole potential, formed by the inner escaping electron and the core. The resulting energy dependence is described by a modulated quasilinear law,

$$\sigma \sim EM(E)/(\ln E - X)^2 , \tag{1a}$$

where E is the energy above the threshold, X is a constant between 4 and 5 (in eV units), and $M(E)$ is the modulation factor,

$$M(E) = 1 + d \sin(\alpha \ln E + \mu)/(\alpha^2 + 1)^{1/2} , \tag{1b}$$

where d is of the order of unity and $\alpha \sim 10$ (Ref. 9). The essential characteristic of this threshold law is the presence of oscillations of varying frequency and amplitude about a gradually increasing average cross section above threshold. However, using the same physical picture and multichannel quantum defect theory, Greene and Rau[10] derived a different behavior in which the amplitude of these oscillations is exponentially small in the dipole moment, and the cross sections would be effectively linear in energy without any observable oscillations above the threshold. Until now, the disagreement between these different theoretical approaches has not been completely settled although the balance of evidence seems to favor that of Wannier.

The first substantial experimental evidence of the validity of the Wannier law was obtained by Cvejanović and Read[11] who deduced $m = 1.131 \pm 0.019$ from the electron-impact ionization of He between 0.2 and 1.7 eV above the threshold. Their indirect result depends on an assumed flat distribution in energies of each of the ejected electrons, predicted by Wannier, which was verified from 0.2–0.8 eV in a separate experiment by Cvejanović and Read[11] and subsequently by Pichou *et al.*[12] for the same experimental system.

An alternative experimental approach to this threshold problem, with much higher energy resolution and simpler final-state configuration, is the study of the two-electron photoionization of atomic negative ions. The first such experiment was performed on H^- by Donahue *et al.*[13] with the use of relativistically Doppler-shifted laser photons. Their results disagreed with the linear law[14,15] while they were fitted equally well, within the uncertainties, to both the Wannier and modulated quasilinear threshold laws. However, in their experiment, the energy-dependent background from a process of photodetachment plus field ionization[13] obscured the actual onset and the threshold behavior of the two-electron photoionization.

Our first experiment was performed[16] on He^- with uv photons generated by a pulsed Nd:YAG (yttrium aluminum garnet) laser. In that experiment details of the threshold behavior of the two-electron photoionization were obscured by uncertainties due to a large background from a two-step two-photon process (photodetachment-photoionization sequence).[16] However, the experiment provided the first absolute cross sections for two-electron photoionization and a determination of the branching ratio between the ejection of two electrons compared to the single electron near the two-electron photoionization threshold.

Most recently, to discriminate between Wannier's and Temkin's theories, Kelly *et al.*[17] searched for the existence of oscillations in the spin dependence of the electron-impact ionization of Na. Their result showed no statistically significant oscillations, contrary to a prediction of Temkin,[8] but in agreement with the Wannier theory. In a later theoretical development of his theory,[9] Temkin incorporated more parameters into his earlier formula and showed that his theory gives a fit to their data that was as consistent as the straight-line fit of the Wannier theory. We note that Greene and Rau's recent theory on Coulomb-dipole picture[10] would also give a straight fit with no observable oscillations.

In this paper we present the result of the near-threshold measurement of K^- two-electron photoionization cross sections. In this case the two-photon two-step ionization is suppressed compared to that in He^- because the "Cooper minimum" in K(4*s*) photoionization[18] cross sections near the employed photon energies greatly reduces the efficiency of the second step. Also, to minimize complications from the photoionization-field-ionization process, we reduced to ~ 300 V/cm the strength of the electric field used to separate product K^+ ions from the K^- beam. This field strength can ionize only Rydberg neutral atoms with $N \geq 30$. Therefore, the background due to this process was negligibly small compared with that of our previous experiment[16] and to that of Donahue *et al.*[15]

II. EXPERIMENTAL DETAILS

Figure 1 shows the main part of the experimental arrangement used in this work. K^- ions were formed in a Colutron ion source[19] by introducing KBr through a movable probe, with no supporting gas for the discharge. The ions were accelerated to 3.1 keV, their momentum was analyzed, then they were directed to a quadrupole deflector *Q*1, whose ion-optical properties and geometry facilitate a coaxial laser–ion beam interaction.[20] The directed ions were deflected through 90° by *Q*1 and then they traversed a 14-cm field-free laser–ion beam interaction region between *Q*1 and a second deflector *D*1, which directed product K^+ ions to an electron multiplier (CEM) and K^- ions to a Faraday cup (FC). The deflector *D*1 was configured to reduce the maximum electric field strength required for the deflection, as mentioned above. A typical K^- current measured at FC was 1 nA. To measure the detection efficiency of K^+ ions, we monitored the signals from collisional detachment at various high negative voltages applied on the CEM cone while maintaining the same voltage difference across the CEM. The results showed a flat plateau above 3.0 kV (where the incident K^+ energy exceeded 6 keV), indicative of 100% detection efficiency. The cone voltage was set at -3.4 kV throughout this experiment. Two 2-mm diameter apertures at the exit of *Q*1 and in front of *D*1 defined the interaction region, as both laser and ion beams initially covered these apertures uniformly and were nondivergent, and thus overlap uniformly over the interaction region. The Doppler shift of the counterpropagating laser beam is ~ 2 meV and was included in determining the photon energies.

The required tunable laser wavelengths between 243 and 260 nm were generated by a Nd:YAG pumped dye laser system with either DCM or LDS 698 dye by doubling the dye-laser output, then mixing the doubled output with the Nd:YAG fundamental. The laser was operated at its highest repetition rate of 17 Hz to suppress the two-photon process by lowering the peak power without significantly reducing the average output power. The laser power was monitored using a vacuum photodiode whose output current was integrated and recorded by a computer. For each run an integrated time-of-flight spectrum of K^+ ion relative to laser shots was obtained by employing a time-to-amplitude converter and a multichannel analyzer.

The K^+ ions could be produced by the following four processes: (a) the desired one-photon two-electron photoionization, (b) two-electron collisional ionization, (c) a two-step photodetachment photoionization, (d) two-step photodetachment field ionization. The background from process (b) was linearly proportional to the background

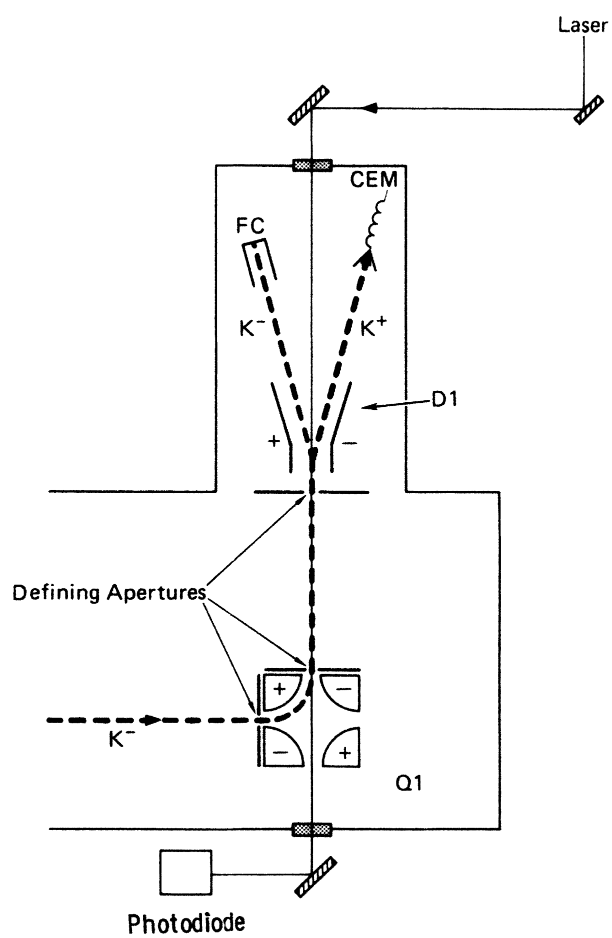

FIG. 1. Schematic diagram of the experimental arrangement.

chamber pressure up to 1×10^{-5} Torr, indicating that process (b) is dominated by a single collisional step at lower pressures. With the operating chamber pressure of $\sim4\times10^{-7}$ Torr and 1 nA of K^- current, the count rate due to process (b) was $\sim3\times10^4$ ions/s.

The single-photon two-electron process (a) to be measured is

$$K^-(4s^2)+h\nu\rightarrow K^+ +2e \ . \tag{2}$$

As both the laser and ion beams filled the beam-defining aperture uniformly, and overlap uniformly over the interaction length l, its cross section $\sigma^{\mp}$ is given by

$$\sigma^{\mp}=Sav/li^-N_p \ , \tag{3}$$

where S is the number of net counts per laser shots after background subtraction, a ($=0.031$ cm^2) is the cross-sectional beam area defined by the apertures, v is the ion velocity, l ($=14$ cm) is the interaction length, i^- is the equivalent K^- beam current (ions/s), and N_p is the number of photons per pulse passing through the defining apetures.

The first step of the two-photon background process (c) is

$$K^-(4s^2)+h\nu\rightarrow K(nl)+e \ , \tag{4a}$$

which is followed by

$$K(nl)+h\nu\rightarrow K^+ +e \ . \tag{4b}$$

This description assumes that the intermediate $K(nl)$ does not radiate during the ~10-ns laser pulse duration. (The shortest-lived state, $4\,^2P$, has a lifetime of 25 ns.) The number of K^+ ions per laser shot from this process (c) is given by

$$S(c)=\left[\sum_{\sigma}\sigma_{nl}^{-0}\sigma_{nl}^{0+}\right]i^-lN_p^2\Big/2a^2v \ , \tag{5}$$

where σ_{nl}^{-0} and σ_{nl}^{0+} are cross sections for (4a) and (4b), respectively. For the photon energies of this experiment, the first step (4a) produces predominantly the lowest channel $K(4s)+\varepsilon p$ products because the first excited channel $K(4p)+\varepsilon s$ dies out rapidly[21] above its threshold at 2.1 eV and all higher-energy channels require two-electron excitation and are relatively suppressed. To our knowledge, no experimental data are available for σ_{2s}^{-0} above 2.7 eV, where it has dropped to about 5×10^{-18} cm^2, and slowly decreases toward higher photon energies.[22] The ionization section σ_{4s}^{0+} has been measured[20] and found to be monotonically increasing from 0.85×10^{-20} to 3×10^{-20} cm^2 as the photon energy increases from 4.77 to 4.98 eV. With $\sigma_{4s}^{-0}\sim10^{-18}$ cm^2, $\sigma_{4s}^{0+}\sim2.5\times10^{-20}$ cm^2, and $N_p\sim1.2\times10^{13}$ (10 μJ/per laser shot), we have $\sigma_{\rm eff}(4s)\sim6\times10^{-24}$ cm^2. This is 10^{-3} times the measured single-photon cross section $\sigma^{\mp}$ at 30 meV above the threshold. To check whether this process contributes significantly to the total counts, we measured the counts as a function of laser power at several different photon energies grater than 50 meV above the threshold. No noticeable nonlinear behavior of the cross sections was observed up to 15 μJ/per laser shot (1.9×10^{13} photons). However, with this small laser power it was difficult to confirm linearity at photon energies within 50 meV above threshold. The actual measurements were made at a laser power corresponding to 10 μJ per pulse.

The background process (d) consists of (4a) followed by field ionization of very-high-n states. These states all have long lifetimes compared with the 1 μs spent by the ions traversing the 14-cm drift space. The maximum electric field used in D1 was ~300 V/cm, which can ionize neutrals produced in reaction (4a) with $n\geq30$. We believe that the contribution from this process is negligible compared with those from process (a) for cross sections greater than 10^{-21} cm^2 which is approximately the lower limit of our observability.

III. RESULTS AND DISCUSSION

Figure 2 shows the cross sections with background contributions mainly from secondary particles created by the scattered laser beam, as the collisional background contribution from the process (b) has been subtracted. We believe the overall background of the measured cross

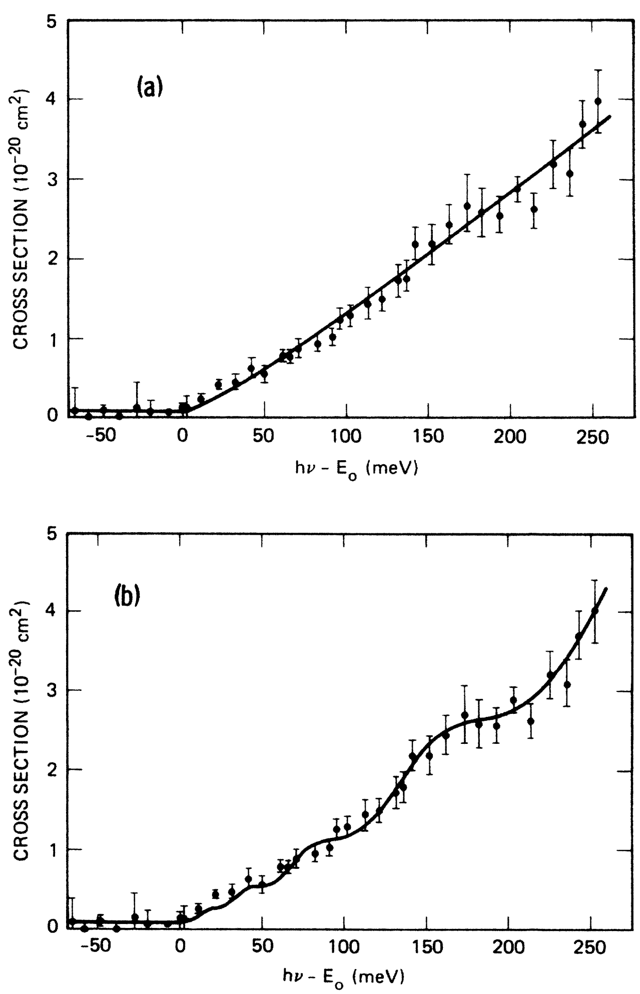

FIG. 2. Absolute cross sections for the K^- double-electron photoionization vs excess photon energy above the threshold. (a) Solid curve is the least-squares fit to a power law. (b) Solid curve is the least-squares fit to a modulated linear law.

sections, including contributions from processes (c) and (d), to be small ($< 10^{-21}$ cm^2) and constant over the energy range studied in this experiment because of the reasons given in Sec. II. The 0 on the abscissa corresponds to the true threshold energy of the two-electron photoionization of K^- [$E_0 = 4.842$ eV, the sum of the ionization potential, 4.341 eV (Ref. 23) and the electron affinity, 0.50147 eV (Ref. 24)]. The energy resolution is ~ 0.5 meV and the total uncertainty in the absolute cross sections is 40%. The error bars correspond to 1 standard deviation from data distributions of several individual runs. The solid line in Fig. 2(a) is the least-squares fit to a power law,

$$\sigma = a_1 + a_2 E^m \ , \tag{6}$$

where σ is in units of 10^{-21} cm^2 and E is in units of meV. The fit results were $a_1 = 0.71 \pm 0.17$, $a_2 = 0.059 \pm 0.014$, and $m = 1.16 \pm 0.05$, in good agreement with Wannier's theory $m = 1.127$. The reduced χ^2 was 0.69 for 34 degrees of freedom resulting in a 91% confidence level. The data fit to the modulated linear law

$$\sigma = b_1 + b_2 E[\ln(E/1000) - 4.5]^{-2} \times [1 + \sin(\alpha \ln E + \mu)/(\alpha^2 + 1)^{1/2}] \tag{7}$$

yielded $b_1 = 0.84 \pm 0.15$, $b_2 = 5.4 \pm 0.2$ $\alpha = 9.4 \pm 0.5$, and $\mu = -1.8 \pm 2.7$. Here we fixed X and d in the equation to be 4.5 and 1, respectively; the reduced χ^2 for this fit was 0.70 for 33 degrees of freedom giving a 90% confidence level. Considering that Eq. (7) has one more parameter than Eq. (6), one may conclude that the fit to the Wannier theory was slightly better than that to the modulated linear law, but the difference is certainly not conclusive. It might be possible that the apparent fluctuations in the data are the results of nonuniformities in the laser beam due to hot spots, which are expected with the laser system that we used. It is worth noting that the fit to the modulated linear law in Fig. 2(b) is strongly influenced by the large fluctuation near 200 meV and appears to be out of phase with the other fluctuations.

Although the energy dependence of the threshold cross sections provides a test of the two theoretical models, a different and more decisive test might be provided by comparing absolute results of theory and experiment. Only recently has the first theoretical calculation of an absolute cross section been made, which included the effects of correlated motion. In that case, McCann and Crothers[25] treated H^- double photoionization semiclassically using a Wannier-type expansion of the wave functions. They obtained a cross section in the Wannier form

$$\sigma^{\mp} = (3.4 \times 10^{-20}) E^{1.127} \ (cm^2) \ . \tag{8}$$

TABLE I. With $\sigma = CE^{1.127}$, C's for various ions are compared. σ is in units of cm^2 and E is in units of eV.

Ions	C (10^{-20} cm^2)	E_A (eV)	E_0 (eV)
H^- (Theory)[a]	3.14	0.7542	14.360
He^- (Expt.)[b]	5.6	0.0774	4.884
K^- (This work)	17.7	0.50147	4.842

[a]Reference 22.
[b]Reference 16.

This equation yields $\sigma^{\mp} \sim 0.8 \times 10^{-20}$ cm^2 at 300 meV above threshold, which is somewhat smaller, but surprisingly similar to our results[16] for He^- double ionization at the same value of E_0. It is also interesting that their formula predicts a ratio $\sigma^{\mp}/\sigma^{-0}$ of the single-photon double detachment (ionization) to single detachement of $\sim 3 \times 10^{-3}$ for H^- at this E, which is essentially the same as we found for He^- ($3.4 \pm 1.2 \times 10^{-3}$) (Ref. 16). Because the species are different, the close agreement is spurious, but the fact that the magnitudes are similar indicates that the theory is reasonable. We can compare our results for He^- and K^- to their calculations by fitting our data to

$$\sigma = CE^{1.127} \tag{9}$$

and obtain the values for C shown in Table I. Also shown in Table I are the threshold photon energies for cross sections σ^{-0} (the electron affinity E_A) and $\sigma^{\mp}$ (the double ionization potential E_0). It is interesting to note that the larger the positive core the larger the C. One might guess there might be some relation between the size of the positive core and C, which is a measure of the absolute cross section.

In conclusion, we have measured the near-threshold absolute cross sections for two-electron photoionization of K^- from 70 meV below to 250 meV above the threshold. The data fit to $\sigma \sim E^m$ yielded $m = 1.16 \pm 0.05$ in agreement with the Wannier theory. However, our result is in similar agreement with Temkin's theory, and thus is still inconclusive in discriminating between the two theories.

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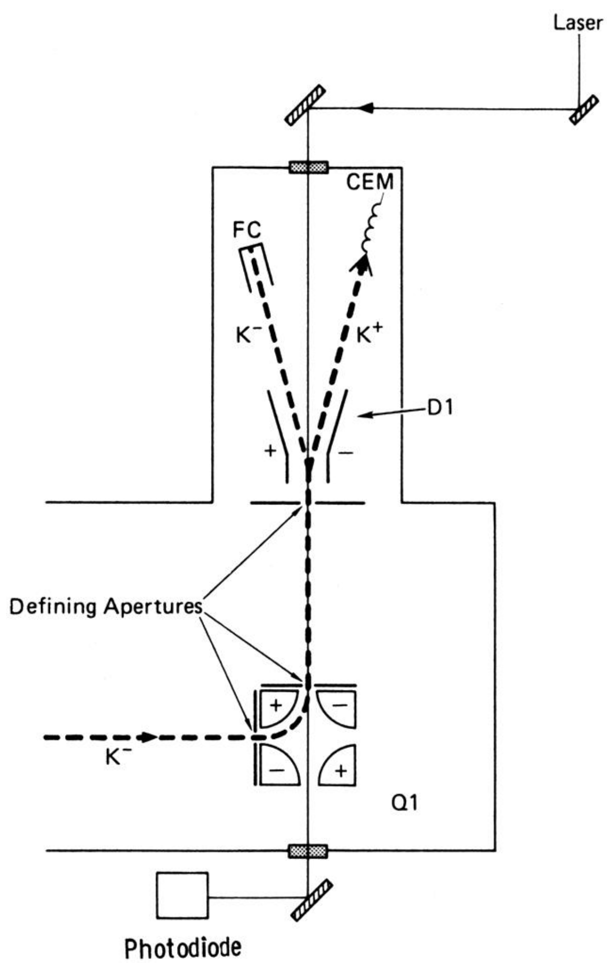

FIG. 1. Schematic diagram of the experimental arrangement.