Angular Correlations in Near-Threshold Double Photoionization of Krypton

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Double ionization of krypton leaving the ion in its $^3P^e$ ground state has been studied at 2.26 eV above threshold, the two photoelectrons being measured in coincidence with angle and energy selection. The results are compatible with the angular dependence of the favored $^3P^o$ state of the pair of electrons, which can be selected by maintaining the angle between the two detectors, θ_{12}, equal to 180°. The observed angular correlation is maximum at $\theta_{12}=180°$ and appears to originate from the three-body ion-electron-electron interaction, in qualitative agreement with the Wannier theory.

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The double ionization of an atom by a single photon is one of the fundamental processes of atomic dynamics. Unfortunately its probability is usually very small, especially in the vicinity of the lowest thresholds where it is not enhanced by indirect and resonant mechanisms, such as the Auger decay, for instance. This is probably the reason why it is still not well understood, and why different models have been proposed so far. Basically the two electrons can be treated on equal footing, and the direct double ionization is then seen as a consequence of their initial correlation. Alternatively, a two-step process [1] can be invoked where a first ionized electron interacts and ionizes the residual singly charged ion. In any case our understanding would be helped by a detailed experimental measurement of the two outgoing electrons.

In previous studies [2–5] integral and singly differential cross sections (with respect to the energy sharing between the two electrons) have been measured. The present Letter reports an experiment in which the energies E_1,E_2 and the *angles* of the two outgoing electrons have been selected for the first time. Therefore, whatever the mechanism responsible for double ionization may be, the observed angular signature should make its identification possible. This is illustrated, for instance, by the model first introduced by Wannier [6], which predicts an angular correlation between the two low-energy electrons escaping from the positive ion with a maximum when their mutual angle θ_{12} is equal to 180°. Previous tests of such a prediction have already been performed in $(e,2e)$ experiments [7,8] but they are partly inhibited by the mixing of numerous final states. From this point of view the double photoionization is much more favorable because of its high final-state selectivity. Furthermore, even within the Wannier model some questions which concern the angular correlation have been raised by many theoretical studies [9–13]. Whereas they all confirm the $E^{1.056}$ threshold law for the total cross section, in agreement with experiment [3], they diverge in the evaluation of the angular correlation, particularly with regard to the dependence of the latter on the ionic charge Z and the specific process considered [13]. For all these reasons double-photoionization measurements including angular information are urgently required.

The apparatus is derived from our low-energy $(e,2e)$ spectrometer [8]. The selector yielding the electron beam has been replaced by a discharge lamp which produces a continuous photon beam. When helium is used, two main spectral lines, namely, He I (21.2 eV) and He II (40.8 eV) contribute to the beam, with intensities of about 75% and 25%, respectively. This beam is crossed at a right angle with an effusive gas beam and the two electrons are detected in coincidence in the two analyzers. The following angular conventions are used: The z axis of the laboratory frame is taken along the unpolarized photon beam, and the two electrons are located by their spherical angles (θ_1,φ_1) and (θ_2,φ_2). Because of cylindrical symmetry around the photon beam the differential cross sections only depend on the azimuthal angles through $\varphi_2-\varphi_1$, and φ_1 is arbitrarily taken to be zero. Although the following results are restricted to the coplanar geometry defined by $\varphi_2=0$ or 180°, the lamp can also be moved out of the plane which contains the two analyzers.

The He I line is convenient for calibration purposes using the single-photoionization process. On the other hand, the double photoionization in the case of krypton can only be achieved by the 40.8-eV photons and an attenuation of the 21.2-eV photons then improves significantly the final true/random coincidence ratio. To this end a filtering system is used. It consists essentially in a disk with 24 holes, which can be rotated by means of a microstepping motor under computer control. A thin film of Formvar is formed on this disk, with transmission coefficients of about 0.1 and 0.6, respectively, for the 21.2- and 40.8-eV photons which pass through one of the holes. The disk is rotated periodically because of contamination by the lamp. Under these conditions a true/random coincidence ratio of about 20 has been achieved in most of the experiments presented below.

We have considered the double-photoionization process

$$h\nu(40.8\text{ eV})+\mathrm{Kr}(4p^6\,{}^1S^e)\rightarrow \mathrm{Kr}^{2+}(4p^4\,{}^3P_2^e)+e+e$$

for which the total energy of the two outgoing electrons $E=E_1+E_2$ is 2.26 eV. The two analyzers were both adjusted to detect electrons of 1.13 eV, with a resolution of

about 0.3 eV (FWHM). The measured true coincidence count rate was typically a few 10^{-3} count/s, and each scan over three or four different angles took several days to reach a statistical accuracy better than 20%. The results are reported in Figs. 1 and 2. In Fig. 1 the mutual angle θ_{12} between the two analyzers has been kept constant and equal to 180°: The triple differential cross section (TDCS) is shown as a function of the single θ_1 angle, after normalization to unity at $\theta_1=90°$. In Fig. 2 the angle θ_1 was kept equal to 90°, 125°, and 150°, successively: The TDCS is plotted in the usual polar-coordinate representation, with a unique scale for the three cases. Small values of θ_2 ($\leq 15°$) could not be attained, due to the proximity of the photon beam. In contrast with a recent double-ionization experiment by high-energy electron impact [14], the two slow photoelectrons appear to be highly anisotropic, both with respect to the beam direction and with respect to each other. To investigate such a behavior we shall use our previous theoretical analysis [15]. Briefly an LS conservation scheme is used, according to which only two states ($^3P^o$ and $^3D^o$) of the pair of electrons can be formed in the present process. Each of these states has a specific angular structure and amplitude. Applying Eq. (12) of Ref. [15] the TDCS can be written as

$$\frac{d^3\sigma}{dE_1\,d\Omega_1\,d\Omega_2}=\sum_{M,M_i,|M+M_i|=1}|\langle 1M_i1M|1(M+M_i)\rangle a_P A_{1M}(\theta_1,\theta_2,\varphi_2) +\langle 1M_i2M|1(M+M_i)\rangle a_D A_{2M}(\theta_1,\theta_2,\varphi_2)|^2 G(180°-\theta_{12}), \quad (1)$$

provided that the two electrons have the same final energy ($E_1=E_2$). So far the only assumption which is made is that the P- and D-state amplitudes have the same θ_{12} dependence, which factorizes out and is accounted for by $G(180°-\theta_{12})$. The physical reason is that the behavior of the cross sections close to threshold is determined by the potential of longest range, as shown by Wigner [16]. In the present case the Coulomb interaction between the two electrons and the ion dominates the centrifugal potential terms and should impose a common dependence. On the other hand, the functions A_{1M} and A_{2M} result from pure angular algebra and are given in explicit form in Ref. [15]. Further application of the Wannier theory gives the energy dependence of a_P and a_D, gives the shape of the G function, and extends the validity of (1) for $E_1\neq E_2$, but we do not use such information for the present work. Thus we treat a_P and a_D as unknown parameters and G as an unknown function.

As the $^3D^o$ state is unfavored [17,18] the A_{2M} angular functions have a node at $\theta_{12}=180°$. In this case Eq. (1) reduces to [15]

$$\frac{d^3\sigma}{dE_1\,d\Omega_1\,d\Omega_2}(\theta_{12}=180°)\propto 1+\cos^2\theta_1. \quad (2)$$

This can be considered as a pure angular algebra result. Thus the agreement with the measurements which ap-

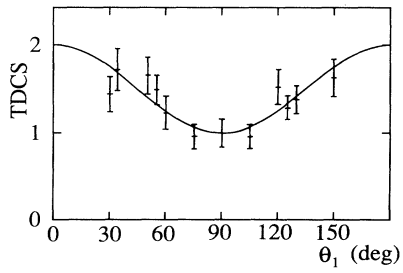

FIG. 1. Measured TDCS for a fixed angle $\theta_{12}=180°$ between the two analyzers. The error bars give the statistical accuracy, and the theoretical TDCS [from Eq. (2)] is shown by the solid line.

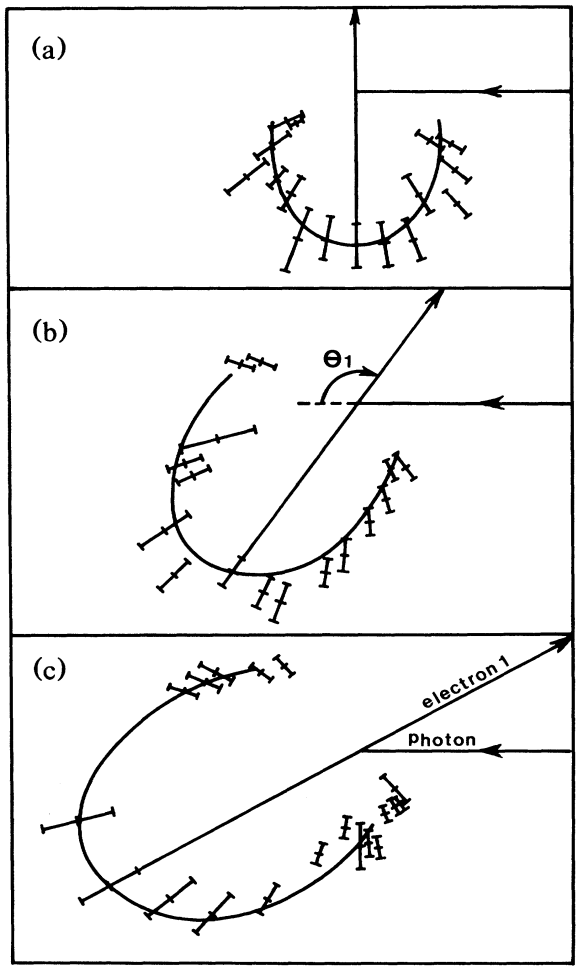

FIG. 2. Measured TDCS for fixed positions of the first electron: (a) $\theta_1=90°$, (b) $\theta_1=125°$, (c) $\theta_1=150°$. The incoming photon beam and first electron are indicated by arrows, and the polar representation with a normalization which is common to (a)–(c) is used.

pears in Fig. 1 does not contain any real physical information, but is a very good confidence test of the full experimental procedure. Of course, the individual angular response of each analyzer has been checked separately against a few known anisotropy parameters β for single photoionization. This indicates that the collisions volume as defined by the intersection of the photon beam, gas beam, and acceptance of one analyzer is constant under rotation of that analyzer. But the agreement of Fig. 1 proves that the same conclusion holds when the acceptance and rotation of the second analyzer are added, which is a sensitive test of the experiment at the TDCS level. More generally the property (2) could be useful to calibrate any type of TDCS measurements in double photoionization.

Now looking at Fig. 2 the main features are the following: (i) For $\theta_1=90°$ the measured TDCS is roughly symmetric with respect to $\theta_2=90°$ or equivalently $\theta_{12}=180°$. (ii) In all cases the maxima of the TDCS are close to $\theta_{12}=180°$, so that an angular correlation between the two electrons clearly appears by scanning θ_1. (iii) For $\theta_1=125°$ and 150° the TDCS become increasingly asymmetric with respect to $\theta_{12}=180°$, despite the lack of data in the region of small θ_2.

Observation (i) is not surprising as in this particular case both the electric field associated with the incoming photon and the first electron are contained in a plane which is perpendicular to the photon beam. Thus the emission diagram for the second electron has to be symmetric with respect to this plane, that is, to $\theta_2=90°$ when restricting to the coplanar geometry. Incidentally it can easily be verified that expression (1) satisfies this property. On the other hand, observation (ii) relies necessarily on the favored [17,18] $^3P^o$ state, which has an antinode at $\theta_{12}=180°$. At first sight it seems to support the Wannier theory, but such a statement requires a more careful examination. Equation (1) shows that the TDCS is a product of two terms of different nature, the first one resulting from angular algebra and symmetry and the second one being the signature of the interaction itself. Now the former already contains a certain amount of angular correlation, which can be labeled as Pauli correlations [19]. This can be illustrated by the example of the A_{10} function [15],

$$|A_{10}(\theta_1,\theta_2,\varphi_2)|^2=2(\cos\theta_1-\cos\theta_2)^2,$$

whose maxima are found when $\cos\theta_1$ and $\cos\theta_2$ are of opposite signs: Even in the independent-particle model two electrons in a $^3P^o$ state are repelled from each other because of the Pauli principle. Thus the help of Eq. (1) is needed for a better understanding of observations (ii) and (iii), and to determine the remaining "interaction-dependent" angular correlation.

First of all, the experimental and theoretical normalizations can be made consistent by setting $a_P=1$ and $G(0)=1$ so that the common value of the TDCS is 1 at the point $\theta_1=90°$, $\theta_{12}=180°$. Then the theoretical TDCS in (1) contains two unknown parameters, $\rho=|a_D|$ and $\alpha=\arg(a_D)$, together with the unknown function G. The results of systematic tests where these two parameters were varied can be summarized as follows.

(1) Excellent agreement with experiment is obtained with the $^3P^o$ state ($\rho=0$) alone. By grouping the measurements into sets of data where θ_{12} is constant (eleven sets for $110°\leq\theta_{12}\leq250°$ have been used), the corresponding G function (G_1) can be deduced point by point, as reported in Fig. 3.

(2) Increasing admixtures of the $^3D^o$ state ($\rho\neq0$) are acceptable provided that it interferes constructively with the $^3P^o$ state, which means $0<\alpha<\pi/2$ rad. This condition is necessary to account for the observed asymmetry (iii), and for the observed width of the lobe at $\theta_1=90°$.

(3) Within the above constraint the main effect of the $^3D^o$ state is to enlarge the lobes. This can be balanced by G functions which are more and more peaked around $\theta_{12}=180°$; the best one (G_2) when taking, for example, $\rho=3$, $\alpha=1$ rad is reported in Fig. 3.

The TDCS calculated with $\rho=0$ and $G=G_1$ have been reported in Fig. 2 (solid line). A smooth curve has been drawn through the points which correspond to the discrete values of θ_{12} (Fig. 3). The curves (not shown) which are obtained with $\rho=3$, $\alpha=1$ rad, and $G=G_2$ are close to the previous ones, and agree with the experimental data as well. This proves that the present set of coplanar data is not yet sufficient for a unique determination of the G function. However, it can easily be verified by means of Eq. (1) that if the $^3D^o$ state starts to interfere constructively when leaving the $\theta_1=90°$, $\theta_{12}=180°$ configuration within the coplanar geometry (by varying θ_2 and keeping $\varphi_2=180°$), then it must interfere destructively along the out-of-plane "equatorial geometry" (keeping $\theta_2=90°$ and varying φ_2 away from 180°). As a consequence, the out-of-plane TDCS calculated in the two above situations ($\rho=0$ and $\rho=3$, $\alpha=1$ rad) differ significantly from each other and measurements in these

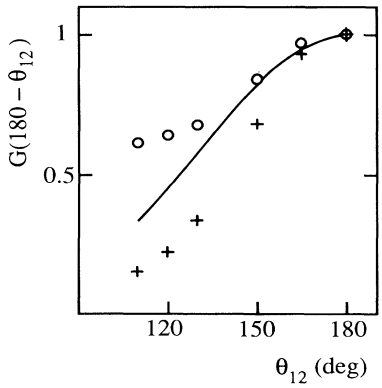

FIG. 3. Correlation factor $G(180°-\theta_{12})$ as deduced from the measurements of Fig. 2, by using Eq. (1): o with $\rho=0$ (G_1) and + with $\rho=3$, $\alpha=1$ rad (G_2). The solid line is a Gaussian with FWHM = 111.6°, from Ref. [10].

conditions should allow a much more precise discrimination in the future.

Returning to the present coplanar measurements, a fundamental result is already obtained. The function G_1 reported in Fig. 3 is the upper limit of the angular correlation function compatible with the results of Fig. 2. Thus an angular correlation which finds its origin in the interaction between the three particles is already established. It appears to be consistent with the Wannier theory, being maximum at $\theta_{12}=180°$. As an illustration, the Gaussian shape of the angular correlation as obtained by Feagin [10], with FWHM $=111.6°$ in the present case, has been plotted in Fig. 3. So far it appears compatible with the present results, being encompassed by the G_1 and G_2 curves.

Such a consistency with the Wannier theory indicates that direct double photoionization is likely to be the dominant process in the present experiment. A similar conclusion has also been drawn by Price and Eland [4] in the case of xenon, on the basis of energy spectra and for equal sharing of the excess energy ($E_1=E_2$). Indirect double photoionization can first proceed through a superresonance, associated with an excited state of the neutral atom lying exactly at the photon energy. This eventuality seems excluded for both xenon and krypton at a 40.8-eV (He II) photon energy, according to the systematic absorption measurements of Codling and Madden [20]. On the other hand, any excited state of the singly charged ion which lies between the $^3P_2^e$ lowest double-ionization threshold and the photon energy can also be responsible, via autoionization, for indirect double photoionization. The best candidates in the present case would be the Rydberg states of Kr^+ with configuration $4p^4(^1D^e)nl$ converging to the $^1D^e$ state of Kr^{2+}. Price and Eland [4] have found indications that such states contribute in the case of xenon. But their participation seems to be minimum for $E_1=E_2$. Moreover, the Rydberg electron in orbital nl would be highly correlated to the first ionized electron as first suggested by Fano [21] and illustrated by experiments where a double-ionization threshold is approached from below [22]. But it is likely that further interaction with the core $4s^24p^4(^1D^e)$ leading to the final $4s^24p^4(^3P^e)$ state of Kr^{2+} would affect the final result of such a correlation. Therefore the pronounced angular correlation which is observed in the present experiment, with a maximum at $\theta_{12}=180°$, suggests that double photoionization is the main process. This is also confirmed by energy spectra which have been taken using a single analyzer and an energy resolution of about 0.1 eV. In the region of double photoionization ($0\leq E_1\leq 2.26$ eV), such spectra exhibit a smooth continuum without any discrete structure that would be the signature of autoionizing states of Kr^+.

To summarize, the present coplanar measurements constitute a first attempt to probe the double-photoionization process in detail. They have been performed in the most favorable case, as the $^3P_2^e$ threshold authorizes the $^3P^o$ favored state for the two outgoing electrons, with an angular form factor which dominates over other states [15]. At this stage it can be claimed that an angular correlation resulting from the three-body interaction has been detected and that it is in qualitative agreement with the Wannier theory. Further investigation will be focused on equatorial-geometry measurements, which should allow an estimation of the $^3D^o$ state contribution and therefore a more precise determination of the G function, as emphasized above. Improvement of the experimental technique should also be possible through multidetection devices, allowing the simultaneous acquisition of data for different angles and/or energies. We have undertaken the building of such an apparatus, which should also make the present type of experiments feasible with other photon sources, like synchrotron radiation.

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[1] J. A. R. Samson, Phys. Rev. Lett. **65**, 2861 (1990).
[2] P. Lablanquie, J. H. D. Eland, I. Nenner, P. Morin, J. Delwiche, and M. J. Hubin Franskin, Phys. Rev. Lett. **58**, 992 (1987).
[3] H. Kossmann, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1226 (1988).
[4] S. D. Price and J. H. D. Eland, J. Phys. B **22**, L153 (1989).
[5] P. Lablanquie, K. Ito, P. Morin, I. Nenner, and J. H. D. Eland, Z. Phys. D **16**, 77 (1990).
[6] G. H. Wannier, Phys. Rev. **90**, 817 (1953).
[7] S. Cvejanovic and F. H. Read, J. Phys. B **14**, 1841 (1974).
[8] P. Selles, A. Huetz, and J. Mazeau, J. Phys. B **20**, 5195 (1987).
[9] A. R. P. Rau, Phys. Rev. A **4**, 207 (1971); J. Phys. B **9**, L283 (1976).
[10] J. M. Feagin, J. Phys. B **17**, 2433 (1984).
[11] F. H. Read, J. Phys. B **17**, 3965 (1984).
[12] C. Bottcher, Adv. At. Mol. Phys. **25**, 303 (1988).
[13] I. Gailitis and R. Peterkop, J. Phys. B **22**, 1231 (1989).
[14] A. Lahman-Bennani, C. Dupré, and A. Duguet, Phys. Rev. Lett. **63**, 1582 (1989).
[15] A. Huetz, P. Selles, D. Waymel, and J. Mazeau, J. Phys. B **24**, 1917 (1991).
[16] E. P. Wigner, Phys. Rev. **73**, 1002 (1948).
[17] A. D. Stauffer, Phys. Lett. **91A**, 114 (1982).
[18] C. H. Greene and A. R. P. Rau, Phys. Rev. Lett. **48**, 533 (1982); J. Phys. B **16**, 99 (1983).
[19] C. D. Lin, Phys. Rev. A **26**, 2305 (1982).
[20] K. Codling and R. P. Madden, J. Res. Natl. Bur. Stand. **76A**, 1 (1972).
[21] U. Fano, J. Phys. B **7**, L401 (1974).
[22] P. A. Heimann, U. Becker, H. G. Kerkhoff, B. Langer, D. Szostak, R. Wehlitz, D. W. Lindle, T. A. Ferrett, and D. A. Shirley, Phys. Rev. A **34**, 3782 (1986).