Fully Differential Cross Sections for Double Photoionization of He Measured by Recoil Ion Momentum Spectroscopy

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(Received 26 March 1996)

Recoil momentum spectroscopy has been used to measure fully differential cross sections for double photoionization of He at energies of 1–80 eV above threshold. The measurement technique allows the determination of the vector momenta of all three escaping particles, and samples the entire final momentum space. When presented in terms of the momentum vector of the center of mass of the electron pair and the relative momentum of the two electrons, the data reveal simple features of the correlated motion. The resulting patterns near threshold are found to be in agreement with a Wannier description of the process. [S0031-9007(96)00803-4]

PACS numbers: 32.80.Fb

The slow breakup of a bound system into three charged particles remains one of the most fundamental and intriguing problems in physics. The final state wave function must represent a high degree of few-body Coulomb correlation involving the simultaneous interaction of all three particles, and no single consensus has appeared on how best to represent this wave function. The case of double photoionization of He is particularly well suited to the study of this problem because (i) the energy delivered to the system can be very well controlled, (ii) the angular momentum delivered to the system through the electric dipole operator is known, and (iii) the ion remaining after the double photoionization is a bare nucleus possessing no relevant internal structure. The best tests of this correlation-dominated process are provided by experiments in which the vector momenta of all three final particles are determined in coincidence. In 1993 Schwarzkopf *et al.* [1] reported the first experiment of this type in which the momentum vectors of the two photoelectrons were detected in coincidence for equal excess-energy sharing. Several subsequent experiments of this type have been reported, covering both equal and unequal energy sharing of the excess energy but always restricted to a coplanar geometry [2–6]. These data have revealed striking correlation patterns. Though many of the major features of the angular distributions are determined by selection rules [7], the quantitative form of the data provides the desired information on the three body decay dynamics. Parallel developments of the theoretical descriptions of the process have demonstrated an ability to account for many of the main features of these patterns [7–13].

In this paper we present fully differential cross sections for double ionization of He near threshold obtained by measuring the momenta of the He^{2+} recoil ion and that of one of the photoelectrons. Here we understand the word fully to mean that, apart from electron spins, the final state of the system is completely kinematically determined. The data differ from those obtained in coincident photoelectron detection in at least two ways which have more than technical importance. First, our data are taken with two detectors which sample at an excess energy of 1 eV, the complete momentum space (4π solid angle) for the ion and one of the electrons, and are recorded event by event. This means that the final momenta of all three particles are determined for *every double ionization event*, with no necessity to choose *a priori* a particular angle or energy for either electron. Thus the entire final five-dimensional momentum space of the escaping three particles is sampled without prejudice, and the physical process itself determines which parts of this space are the most important. In principle, a similar measurement could be performed by measuring coincident photoelectrons for all polar and azimuthal angles and energy combinations, but this is an experimentally hopeless task with the solid angles of typical electron spectrometers. Second, the recoil ion momentum itself, which is equal and opposite to the center-of-mass momentum of the ejected electron pair, appears to be a particularly convenient coordinate for the description of the physical process at hand.

Thus instead of the conventional momentum coordinates $\mathbf{k}_1$ and $\mathbf{k}_2$ of the two electrons, we choose the (Jacobi) momentum coordinates $\mathbf{k_r} = \mathbf{k}_1 + \mathbf{k}_2$ and $\mathbf{k_R} = \frac{1}{2}(\mathbf{k}_1 - \mathbf{k}_2)$ of the electron-pair center-of-mass (CM) motion and the electron-pair relative motion, respectively [14]. Neglecting the photon momentum, the ion momentum is $-\mathbf{k_r}$. These momenta are just the conjugate coordinates of the relative vectors $\mathbf{r} = \frac{1}{2}(\mathbf{r}_1 + \mathbf{r}_2)$ and $\mathbf{R} = \mathbf{r}_1 - \mathbf{r}_2$ of the electron-pair CM and interelectronic

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axis, respectively. The electric dipole operator is exactly $\hat{\boldsymbol{\epsilon}} \cdot \mathbf{r}$ in these coordinates and hence excites directly the $\mathbf{k_r}$ motion of the He^{2+} relative to the electron-pair CM. Interelectronic excitation along $\mathbf{k_R}$ derives from subsequent coupling to the $\mathbf{k_r}$ motion, which is at the heart of double ionization and the few-body Coulomb problem. Especially near threshold, the use of these relative coordinates reveals naturally certain simple characteristics of the strongly correlated motion of the electron pair not easily seen in the $\mathbf{k}_1, \mathbf{k}_2$ description. As has been pointed out on several occasions previously, this situation is similar to that encountered in nuclear physics, where the excitation of rotational and vibrational motion is better described in terms of collective rather than single-particle coordinates.

The use of cold target recoil momentum spectroscopy (COLTRIMS) in both ion-He and photon-He collisions is now well established, and is described in Refs. [15–18]. The essential feature of the method is that the He target is cooled in a supersonic jet to an internal temperature below 0.2 K, for which its thermal momentum (<0.05 a.u.) is much less than the momentum transfer which the ion receives in the physical process of interest (typically >1 a.u.). In the version of the apparatus we have used for the present study, the cooled gas jet was crossed by a photon beam from the Advanced Light Source (ALS) at LBNL and a weak transverse electric field (2 V/cm) was used to accelerate the He ions onto the surface of a channel plate Z stack equipped with a position-sensitive wedge-and-strip anode. The ALS was operated in a two-bunch mode which allowed the time of flight of the recoil ion to be measured with a precision near 1 ns. The vector momentum of the He ion was determined from a combination of this time of flight and the position at which the ion struck the detector. A second position-sensitive detector located facing the ion detector but on the opposite side of the interaction region detected one of the photoelectrons in coincidence with the recoil ion. The vector momentum of this photoelectron was calculated in the same way as was that of the recoil ion. Events in which both photoelectrons struck the electron detector, which occurred with low but nonzero probability, were rejected based upon the pulse-height distribution from the electron detector. Our recoil ion detector had 4π solid angle for all excess energies (E_{exc}). The electron detector, however, had a full solid angle only up to $E_{\mathrm{exc}} =$ 1 eV. Therefore momentum distributions at $E_{\mathrm{exc}} =$ 1 eV are measured with an electron-recoil coincidence, yielding the $\mathbf{k_r}$ and $\mathbf{k_R}$ distributions, while for the higher energies only the recoil momentum distributions could be determined in coincidence with the machine pulse.

In Fig. 1 we present density plots of the momentum distributions measured for a photon energy 1 eV above the double ionization threshold. The distributions show projections of all events with $-0.1 < k_x < 0.1$ a.u. into the y-z plane, where z lies along the polarization vector of the electric field and the photon propagates in the x direction. Figure 1(a) shows the He^{2+} recoil ion momentum distribution which is equivalent to the $\mathbf{k_r}$ distribution. The distribution appears qualitatively dipolelike in character, even so near threshold. Figure 1(b) shows the distribution of $\mathbf{k}_1$ (or $\mathbf{k}_2$), the laboratory momentum of either electron, and demonstrates that the strong interaction between the electrons has completely removed the simple dipolelike distribution which one sees in single ionization, or in double ionization at photon energies well above threshold. This result is known from previous measurements [19,20]. Figure 1(c) shows the $\mathbf{k_R}$ distribution and hence the relative motion of the two electrons. It indicates that the electron

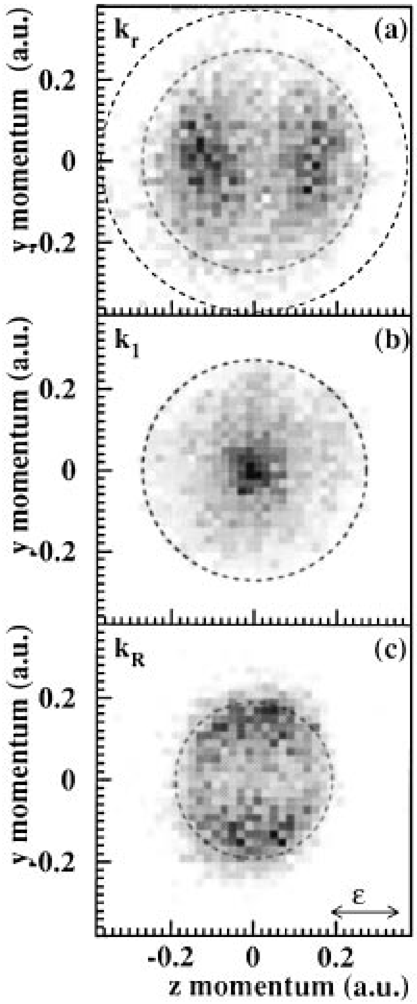

FIG. 1. Density plots of projections of the momentum spectra from double ionization of He by 80.1 eV photons. The z and y components of the momentum are plotted on the horizontal and vertical axes, respectively, the polarization vector of the photon is in the z direction and the photon propagates in the x direction. Only events with $-0.1 < k_x < 0.1$ a.u are projected onto the plane. (a) The He^{2+} recoil ion (or $-\mathbf{k_r}$) momentum distribution. The outer circle indicates the maximum possible recoil ion momentum, and the inner circle is the locus of events for which the $\mathbf{k_r}$ motion has half of the excess energy. (b) The distribution of single electron momenta ($\mathbf{k}_1$ or $\mathbf{k}_2$). The circle locates the momentum of an electron which carries the full excess energy. (c) The relative electron momentum (or $\mathbf{k_R} = \mathbf{k}_1 - \mathbf{k}_2$) distribution. The circle identifies the maximum possible value for $\mathbf{k_R}$.

pair separates preferentially perpendicular to the photon polarization axis and therefore perpendicular to the recoil momentum $-\mathbf{k_r}$. This is qualitatively consistent with a Wannier picture of the process. The He^{2+} ion and the electron pair separate in such a way that the electron-pair CM remains near the ion but stably oscillates perpendicular to the "Wannier ridge" along $\mathbf{R}$, or essentially k_R. Motion of the electron-pair CM along the Wannier ridge is unstable and may result in one or the other of the electrons recombining with the ion, blocking double escape.

In the dipole approximation the momentum dependence of the fivefold differential cross section is characterized by overlap matrix elements $\langle \mathbf{k_r k_R} | \psi \rangle$ of detector states $\langle \mathbf{k_r k_R}|$ with ${}^1P^o$ continuum states $|\psi\rangle$ of the electron pair [14]. These quantities are related to the momentum representation $\tilde{\psi}(\mathbf{k_r}, \mathbf{k_R})$ of the continuum states and are a linear combination $\tilde{\psi} = \sum_K c_K \tilde{\psi}_K^\zeta$ of substates with projection $K = \hat{\mathbf{R}} \cdot \mathbf{L}$ of the total angular momentum along the Wannier ridge and with a definite evenness or oddness along the ridge denoted by the quantum number $\zeta = 0$ or 1. For ${}^1P^o$ states, $K = 0, 1$ and $K + \zeta = 1$. When approximated by Wannier wave functions, $\tilde{\psi}_K^\zeta$ gives in leading order the familiar energy dependence $\sigma \sim E^{m(1+2\zeta)}$ of the total cross section with $m = 1.056$ in He. In the Wannier theory, the mixing coefficients c_K remain undetermined; however, minimal excitation along the Wannier ridge is generally observed both above and below the double ionization threshold. Hence one expects $\zeta = 0, K = 1$ to dominate over $\zeta = 1, K = 0$ excitation. From the experiments performed using traditional coincident electron spectroscopy $(\gamma,2e)$ no pictures equivalent to Fig. 2 can be obtained since they are restricted to selected angular and energy combinations between the electrons. However, Lablanquie *et al.* [5] could show from their $(\gamma, 2e)$ data that at $E_{\text{exc}} = 4$ eV the ratio of $K = 1$ (which is the a_u term in their notation) to $K = 0$ is smaller than $1/16$.

Figures 2(a)–2(d) show $\int |\tilde{\psi}_K^\zeta|^2 \, d\Omega_x$ for $K = 0$ and $K = 1$ calculated using an extended *fourth-order* Wannier description [14] which allows a direct Fourier transform of the Wannier coordinate wave function $\psi_K^\zeta(\mathbf{r}, \mathbf{R})$ and hence direct determination of the momentum wave function $\psi_K^\zeta(\mathbf{k_r}, \mathbf{k_R})$, which is undefined in the second-order Wannier theory. Comparison of Figs. 2(c) and 2(d) with the data shown in Fig. 1(c) clearly shows that a $(\zeta = 0, K = 1)$ excitation is favored.

A more quantitative discussion of these results can be carried out by focusing on the relative partition of the excess energy between $\mathbf{k_R}$ and $\mathbf{k_r}$ motions and on the angular distributions of $\mathbf{k_R}$ and $\mathbf{k_r}$. The total excess energy, which is the difference between the photon energy and the double ionization threshold energy of 79.1 eV, can be written as (in a.u.) $E = \frac{1}{4} k_r^2 + k_R^2$. Figure 3

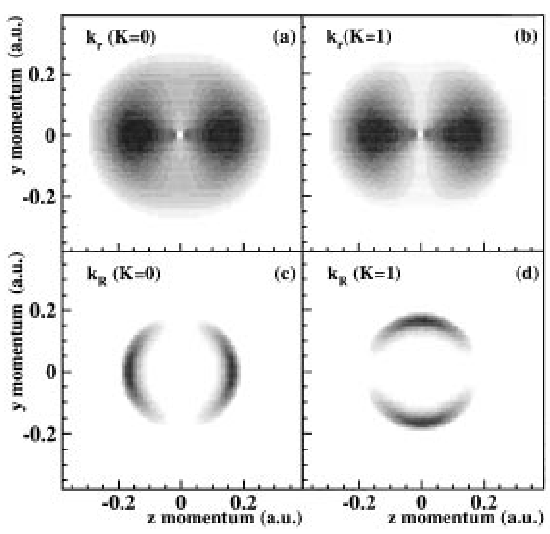

FIG. 2. Density plots calculated from the Wannier model, corresponding to Figs. 1(a) and 1(c), for different choices of the K quantum number (see text).

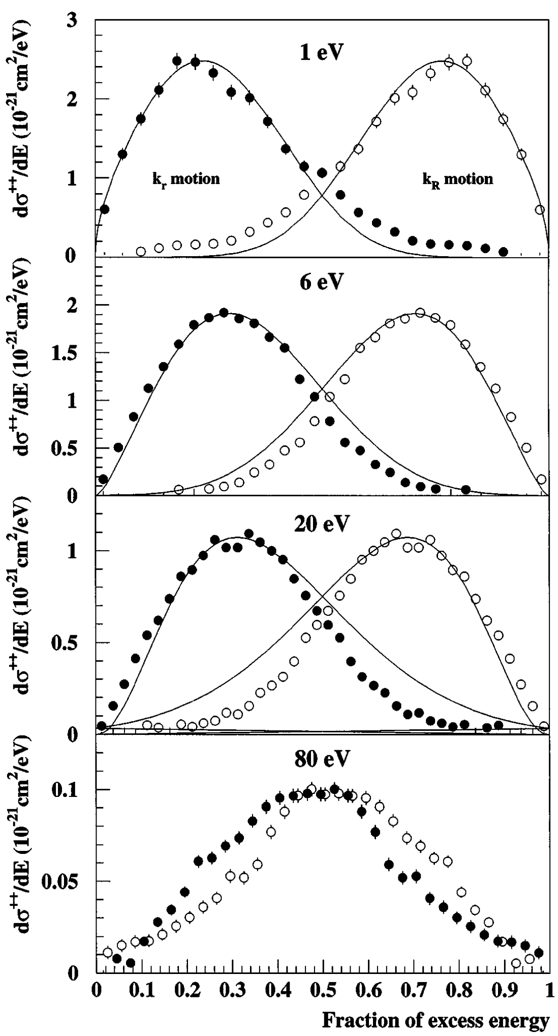

FIG. 3. Cross sections differential in energy plotted as a function of the fraction of the excess energy in the $\mathbf{k_r}$ (filled circles) or $\mathbf{k_R}$ (open circles) motion. The solid curve in the upper three figures is from the fourth-order Wannier calculation. The theoretical curves are normalized to the experiment, which is on an absolute scale.

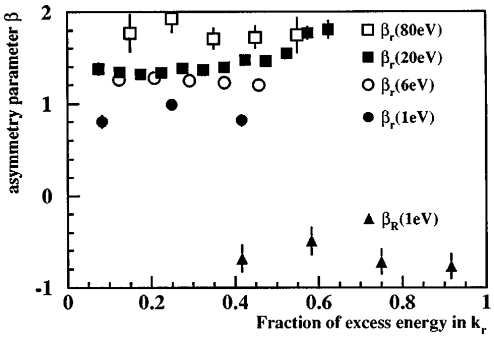

FIG. 4. Experimental asymmetry parameters for the orientation of the interelectron axis ($\mathbf{k_R}$ motion) at an excess energy of 1 eV (full triangles) and of the recoiling nucleus ($\mathbf{k_r}$ motion) for 1 eV (full circles), 6 eV (open circles), 20 eV (full squares), and 80 eV (open squares) as a function of the fraction of the excess energy in the $\mathbf{k_r}$ motion.

shows measured cross sections differential in energy, plotted separately for $\mathbf{k_R}$ and $\mathbf{k_r}$ motion, for several photon energies. Far above threshold the energy sharing is seen to be approximately equal, while near threshold much more energy is carried in the relative motion of the two electrons than by their center of mass motion. The data are well described by $\int |\tilde{\psi}_{K=1}^{\zeta=0}|^2 \, d\Omega_r \Omega_R$ calculated from the fourth-order Wannier theory [14] (solid line) even quite far above threshold. Note that the experimental data are on an absolute scale, with an error of about 7% for the absolute value. They have been normalized to the cross sections of [21] using the ratio of double to single ionization from [17,22]. The Wannier calculation has been normalized to fit the maximum. A recent *ab initio* calculation by Pont and Shakeshaft [23] shows very good absolute agreement with our experimental results for 6, 20, and 80 eV excess energy.

The angular distributions of $\mathbf{k_r}$ (the recoil ion) and $\mathbf{k_R}$ with respect to the polarization axis can be parametrized in the usual way by angular asymmetry parameters β_r or β_R, respectively, where $d\sigma/d\omega \sim 1 + \beta P_2(\cos\theta)$. A straightforward calculation in the fourth-order Wannier theory near the "cold-ion" limit $k_r \rightarrow 0$ (or $k_R \rightarrow \sqrt{E_{\mathrm{exc}}}$) gives for $K = 1$ $\beta_r \simeq \frac{7}{5}$ and $\beta_R \sim eq - 1$; for $K = 0$, it gives $\beta_r \simeq \frac{4}{5}$ and $\beta_R \simeq +2$. Comparison with the 1 eV measured values in Fig. 4 shows that the $K = 1$ excitation is favored, although some $K = 0$ mixing occurs. With increasing photon energy we find $\beta_r \rightarrow 2$. For high photon energies the energy between the two electrons gets asymmetric. In this case the recoil ion will reflect the angular dependence of the fast electron (k_1) and one expects $\beta_r \approx \beta_{k1} \rightarrow 2$.

In conclusion, we have used cold target recoil ion momentum spectroscopy to measure completely differential cross sections for double ionization of He from 1 to 80 eV above threshold. The momentum distribution of the He^{2+} recoils shows a qualitatively dipolelike character at even the lowest photon energy, but quantitatively differs from a pure $\cos\theta^2$ distribution by an amount which proves to be a sensitive probe of theory. The He atom preferentially breaks up such that the nucleus recoils along the photon polarization axis while the two electrons separate along a line perpendicular to the polarization.

This work was supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research, U.S. Department of Energy. The Max Planck Preis of the Alexander von Humboldt Stiftung, the DFG and BMBF, the Graduiertenförderung des Landes Hesses, and the Willkomm Foundation. One of us (R. D.) gratefully acknowledges support from the Alexander von Humboldt Foundation and of the Studienstiftung des deutschen Volkes (V. M.). We thank T. Warwick, E. Rotenberg, J. Denlinger, and the entire support staff of the Advanced Light Source for the extraordinary support which made this experiment possible.

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