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LETTER TO THE EDITOR

Recoil ion and electronic angular asymmetry parameters for photo double ionization of helium at 99 eV

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Abstract. Recoil ion momentum spectroscopy has been used to map the entire five-dimensional momentum space of the photo double ionization of helium at 20 eV above threshold. Angular asymmetry parameters for the relative motion of the electrons and the recoil ion have been determined and are found to be close to similar data at 1 eV above threshold. In addition the asymmetry parameter of one photoelectron is found to be in good agreement with recent theory.

Photo double ionization of helium by a single photon is one of the most fundamental many-body processes and has been widely studied in the past experimentally as well as theoretically. The final-state wavefunction must include the simultaneous interaction of all three particles in the continuum and no single consensus has appeared on how best to represent this wavefunction. On the experimental side, recent advances in synchrotron light sources have made it feasible to measure the vector momenta of all three particles in coincidence. Following the first experiment by Schwarzkopf *et al* [1] several experiments have measured the momentum vector of the two photoelectrons in coincidence, covering both equal and unequal energy sharing of the excess energy but always restricted to a coplanar geometry [2–6].

Fully differential cross sections for the double ionization of helium without any *a priori* restriction to a particular angle or energy for either electron were first measured by Dörner *et al* [7] for an excess energy of 1 eV using recoil ion momentum spectroscopy. Here the word ‘fully’ is understood to mean that, apart from the spins, the final state of the system is completely kinematically determined. That is, the final momenta of all three particles are determined for all angles and energies of the fragments simultaneously. This close to the double ionization threshold, good agreement has been found with a fourth-order Wannier theory by Feagin [8, 9] as well as with *ab initio* calculations by Pont and Shakeshaft [10].

In this paper fully differential cross sections for the double ionization of helium farther away from the threshold (at 20 eV excess energy) are presented. In experiments using coincident electron detection, differences in the shape of the cross section have been found

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between excess energies close to threshold and in the range of 20 eV above threshold at selected electron emission angles and unequal energy sharing [5, 6]. One aim of the present work is to investigate the influence of the excess energy on the fully differential cross section for all angles and energy sharings.

The measurements have been performed at the Advanced Light Source (ALS) at LBNL using the well established method of cold-target recoil-ion momentum spectroscopy (COLTRIMS) [7]. A precooled supersonic helium gas jet with an internal temperature below 0.4 K was crossed with a beam of 99 eV photons, which were linearly polarized with a Stokes parameter of $S_1 = 0.98$. A weak transverse electric field (7 V cm^{-1}) was used to collect all He ions onto a two-dimensional position-sensitive detector with a 4π solid-angle collection efficiency. The two-dimensional position information together with the time-of-flight information allows the calculation of the He ion momentum vector. The photoelectron momenta are obtained in a similar way with a second two-dimensional position-sensitive detector. The recent measurements by Dörner *et al* were restricted to 1 eV excess energy to ensure 4π solid-angle collection efficiency for the electrons. To overcome this restriction in this work a homogeneous 10 G magnetic field along the spectrometer axis was used to trap electrons with higher energy in the spectrometer and guide them to the detector. This technique has already been widely used in conjunction with COLTRIMS in ion–atom collisions [11–14]. Again, from the position on the detector and the electron time of flight the electron momentum vector can be computed. Using momentum conservation, the momentum vector of the second, undetected, electron is computed. Using the magnetic field a 4π solid angle for collection was achieved for electrons with energy $E \leqslant 10$ eV. This in no way restricts our measurement because in the case of double ionization at 20 eV excess energy, one of the two electrons always fulfils the condition $E \leqslant 10$ eV.

The complete determination of the momentum vectors of all three particles in the final state for each event allows transformation into any desired coordinate system. Instead of the conventional electron momenta $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ in the laboratory reference frame, the Jacobi momentum coordinates $\boldsymbol{k_r} = \boldsymbol{k}_1 + \boldsymbol{k}_2$ of the electron-pair centre-of-mass (CM) motion and $\boldsymbol{k_R} = \frac{1}{2}(\boldsymbol{k}_1 - \boldsymbol{k}_2)$ of the electron-pair relative motion [9] have been proven useful near threshold. These momenta are the conjugate coordinates of the relative vectors $\boldsymbol{r} = \frac{1}{2}(\boldsymbol{r}_1 + \boldsymbol{r}_2)$ and $\boldsymbol{R} = \boldsymbol{r}_1 - \boldsymbol{r}_2$ of the electron-pair CM and interelectronic axis respectively.

Figure 1 shows density plots of the momentum distributions for $\boldsymbol{k_r}$, $\boldsymbol{k}_1$ and $\boldsymbol{k_R}$ at 20 eV excess energy. Atomic units are used throughout this report for momenta. The distributions shown are projections of the complete three-dimensional momentum distribution into the y–z plane for events with $-0.2 < k_x < 0.2$, where z lies along the polarization vector $\hat{\boldsymbol{\epsilon}}$ of the photon beam and the photons propagate in the x direction. A similar behaviour is found as in the data at 1 eV above threshold by Dörner *et al* [7]. The $\boldsymbol{k_r}$ distribution (figure 1(*a*)) appears qualitatively dipole-like in character, reflecting the dipole operator $\hat{\boldsymbol{\epsilon}} \cdot \boldsymbol{r}$ acting on the electron-pair CM [9]. Likewise, even at 20 eV excess energy the strong interaction between the electrons still completely removes the simple dipole-like structure in the $\boldsymbol{k}_1$ spectrum seen in single photoionization or at energies further above threshold (figure 1(*b*)) [15]. The relative motion $\boldsymbol{k_R}$ of the electrons in their CM system is shown in figure 1(*c*). It shows the electron pair separating preferentially perpendicular to the photon polarization axis and hence perpendicular to the recoil momentum $-\boldsymbol{k_r}$. This is surprisingly similar to results at 1 eV excess energy. In a Wannier description of the process this orientation of $\boldsymbol{k_R}$ perpendicular to $\hat{\boldsymbol{\epsilon}}$ is directly related to excitation of the Wannier state with $K = 1$, where $K = \hat{\boldsymbol{R}} \cdot \boldsymbol{L}$ is the projection of the total angular momentum $\boldsymbol{L}$ along the interelectronic axis. The second allowed Wannier mode for He photo double ionization with $K = 0$ leads to a separation of the electron pair along $\hat{\boldsymbol{\epsilon}}$ [9, 7].

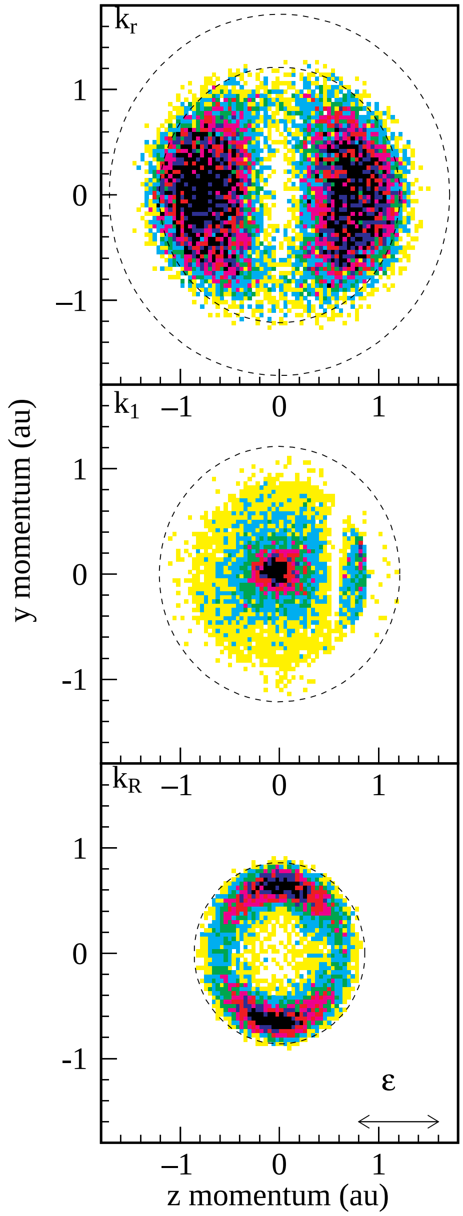

Figure 1. Density plots of the momentum distribution from the double ionization of He by 99 eV photons. The plots are projections onto the y–z plane (y: direction of He jet motion, z: direction of $\hat{\epsilon}$) with $-0.2 < k_x < 0.2$. (a) $\boldsymbol{k_r} = \boldsymbol{k}_1 + \boldsymbol{k}_2$ (recoiling He^{2+}) momentum distribution. The outer circle indicates the maximum possible momentum and the inner circle indicates the momentum for which the $\boldsymbol{k_r}$ motion has half of the excess energy. (b) Single-electron momentum distribution $\boldsymbol{k}_1$. The circle shows the momentum of an electron which carries the full excess energy. (c) $\boldsymbol{k_R} = \frac{1}{2}(\boldsymbol{k}_1 - \boldsymbol{k}_2)$ momentum distribution. The circle shows the maximum value for $\boldsymbol{k_R}$.

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A more detailed discussion can be carried out by using the common parametrization of angular distributions with respect to the polarization axis: $d\sigma/d\Omega \sim 1 + \beta P_2(\cos\theta)$ where β is the angular asymmetry parameter. This parametrization has been widely used to characterize the angular distribution of the photoelectron (β_1) and it has been shown that β_1 depends on the excess energy as well as on the energy of the electron [16, 15]. It can also be used to characterize the angular distribution of the $\boldsymbol{k_r}$ (β_r) and $\boldsymbol{k_R}$ (β_R) motion. The measured β-parameters at 20 eV excess energy and their dependence on the fraction f of the energy carried in the respective motion are shown in figure 2 in comparison to relevant data from Dörner *et al* [7] and Wehlitz *et al* [15].

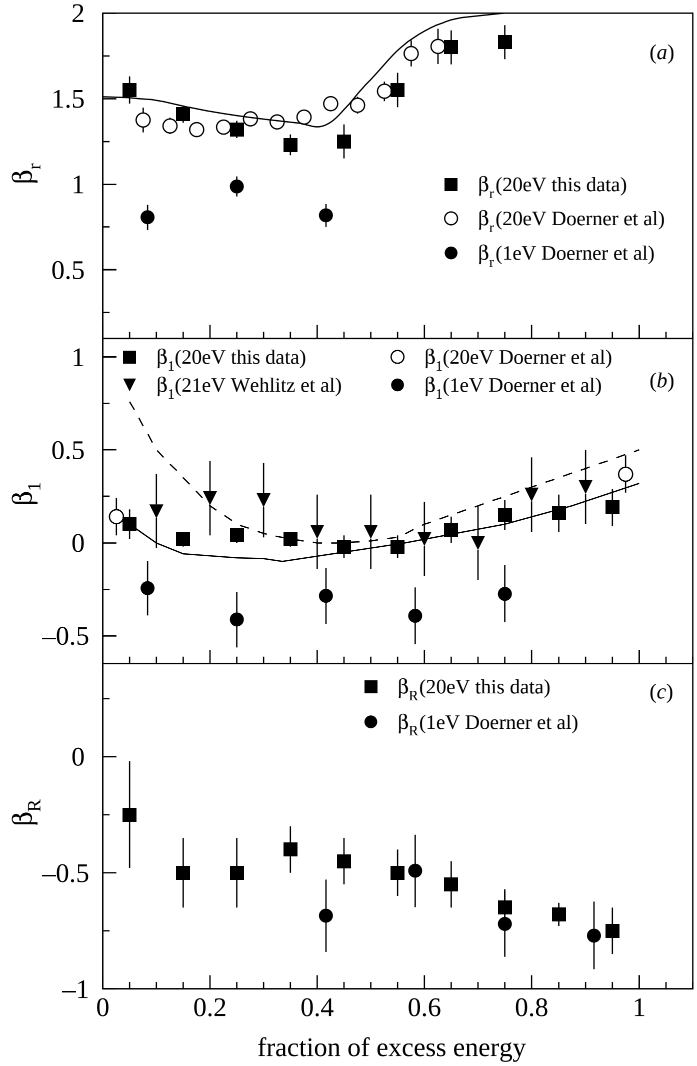

Figure 2. Experimental asymmetry parameters for (*a*) $\boldsymbol{k_r}$, (*b*) $\boldsymbol{k}_1$ and (*c*) $\boldsymbol{k_R}$. The abscissa gives the fraction of excess energy contained in the corresponding motion. Our data are compared with data from Dörner *et al* [7] at 1 eV and 20 eV and for β_1 with data from Wehlitz *et al* [15] at 21 eV. The full curve in (*a*) is a calculation by Pont and Shakeshaft [10]. The full curve in (*b*) represents a calculation by Proulx and Shakeshaft [21] for 21 eV. The broken curve in (*b*) is a calculation by Maulbetsch and Briggs [20] using a final-state wavefunction consisting of the product of 3C wavefunctions for 21 eV excess energy.

β_r (figure 2(*a*)) shows the strong dipole-like distribution of the electron-pair CM motion, i.e. the motion of the recoiling ion, along the polarization axis. The data are in good agreement with the the data of Dörner *et al* at 20 eV, which shows the reproducibility of COLTRIMS experiments. Also shown in figure 2(*a*) is the result of an *ab initio* calculation by Pont and Shakeshaft [10] for 20 eV. They describe the final state by a product of two screened Coulomb (2SC) wavefunctions employing effective charges. A good agreement between experiment and theory is found for the dependence of β_r on the fraction of excess energy. The data clearly show a sharp increase in β_r approaching $\beta_r \approx 2$ for larger f. This can be understood as follows. For $f \approx 1$ almost all the energy is in the CM motion of the electron pair and the relative motion of the electrons is very small. The two electrons emerge as if they were a single particle and the process thus resembles a single photoionization process. In this case there is only one preferred axis, the electric-field axis, and the angular distribution is dipolar, corresponding to $\beta_r = 2$. The extreme case where there is no relative motion of the electrons is, however, suppressed by Coulomb repulsion of the electrons.

For the relative motion of the electrons in their CM system we find a value for β_R which is not different from the values measured at 1 eV excess energy by Dörner *et al* [7]. The better statistics of our experiment allowed for more data points with smaller error bars. β_R seems to decrease slightly with the increase of energy in the relative electron motion. A large fraction of excess energy in the $\boldsymbol{k}_R$ motion corresponds to the electron-pair CM (and the recoil ion) remaining stationary at the point of interaction ('cold-ion'). In this case the data indicate a strong alignment perpendicular to the polarization axis, which becomes more uniform, when most of the energy lies in the CM motion. It should be noted that $\boldsymbol{k}_R = 0$, which corresponds to electrons of equal energy emerging together with no relative momentum, is forbidden by their mutual Coulomb repulsion; the maximum cross section is obtained for $\boldsymbol{k}_R = 70\%$ of its maximum value.

For a pure $K = 1$ excitation the fourth-order Wannier theory predicts $\beta_r \simeq \frac{7}{5}$ and $\beta_R \simeq -1$ near the 'cold-ion' limit ($\boldsymbol{k}_r \to 0$). Comparison with our β_R data shows that, although $K = 1$ excitation is favoured, some $K = 0$ mixing occurs. Extrapolating our data to $\boldsymbol{k}_r = 0$, the ratio c_1/c_0 of the complex amplitudes of the two Wannier states ($K = 1$, $K = 0$) in the 'cold-ion' limit [17] can be calculated. For 20 eV, with $\beta_r = 1.62$ and $\beta_R = -0.76$ we obtain an absolute value $|c_1/c_0| = 2.4$, showing the dominance of the $K = 1$ excitation and the $K = 0$ mixing.

Experiments using electron spectroscopy have studied the asymmetry parameter β_1 of one of the photoelectrons from threshold [18, 19] to excess energies up to 41 eV [15]. Figure 2(*b*) shows our data at 20 eV and data at 1 eV by Dörner *et al* [7] obtained with COLTRIMS in comparison with 21 eV data by Wehlitz *et al* [15]. The data at 1 eV by Dörner *et al* [7] are found to be in very good agreement with the measurements by Hall *et al* [18] and Wehlitz *et al* [15]. The extensive studies of Wehlitz *et al* [15] show the increase of β_1 with excess energy and a change in the dependence of β_1 on the energy sharing. While close to threshold the β_1 curves are flat, they become U-shaped at higher energies. Our data confirm this trend at 20 eV and agree within the larger error bars with the revised data of Wehlitz *et al* [15]. Furthermore our recent data agree well with the only two data points Dörner *et al* [7] could obtain at 20 eV for extremly unequal energy sharing.

Also shown in figure 2(*b*) is a calculation by Maulbetsch and Briggs [20] (broken curve) using a product wavefunction consisting of three two-body Coulomb (3C) wavefunctions in velocity form for the final state. This wavefunction includes electron–electron correlation through factors involving relative momentum $\boldsymbol{k}_R$ of the electrons. The full curve shows calculations by Proulx and Shakeshaft [21] calculated with similar code as the ionic (β_r) distribution (2SC) in figure 2(*a*). The 3C and 2SC calculations are close at equal energy sharing but differ significantly at low energies. Our data clearly confirm the flat shape of β_1 predicted by the 2SC and are for low energies in significant disagreement with the 3C results.

Recent measurements by Schwarzkopf *et al* [2], Lablanquie *et al* [5] and Viefhaus *et al* [22] have concentrated on the effect of energy sharing on the coincident angular distribution of the electrons. All these experiments are restricted to the case where one electron is emitted along the polarization axis. For equal energy sharing, selection rules place firm constraints on the angular distribution of the other electron, resulting in nodes at 180° and 0°. Close to threshold it has been found that the shape of the angular distribution does not vary significantly with the energy sharing. At higher energies, such as 18.6 eV [5], the symmetry contraints are much less rigorous for unequal energy sharing. In this case the node at 180° even turns into a maximum.

The emission of two electrons along the polarization axis corresponds to the $K = 0$ mode in the Wannier theory discussed above. Lablanquie *et al* [5] conclude from their measurements that this mode becomes more pronounced with increasing energy for unequal energy sharing. While the β_r and β_R parameters indicate a certain mixing between $K = 0$ and $K = 1$ states, we see no difference in β_R between 1 eV excess energy and 20 eV. However, electron angular distributions extracted from our data, which will be discussed in detail in a forthcoming paper, also show this filling of the node at 180° for unequal energy sharing. The close agreement in the β_R data for 1 eV and 20 eV, in view of the electron spectroscopy experiments discussed, can be attributed to the fact that the β_R data sample the complete momentum space, whereas the other experiments based on photoelectron spectroscopy focus on a very small subsample of the total cross section, centred in momentum space on a point for which the $K = 1$ mode cannot contribute and only the small $K = 0$ amplitude is visible. They are thus very sensitive to changes in the strength of that mode. The current COLTRIMS data for 20 eV, however, indicate that for the total cross section the $K = 1$ contribution still dominates the $K = 0$ contribution.

In conclusion we have used COLTRIMS to extend the measurements of fully differential cross sections to a photon energy of 99 eV which is 20 eV above the double ionization threshold. The momentum distributions in Jacobi coordinates and the respective asymmetry parameters show a behaviour similar to that seen in previous measurements by Dörner *et al* [7] at 1 eV above threshold. The data are in good agreement with a Wannier picture of the ionization process, where the ion recoils along the polarization axis of the photon and the electron pair separates perpendicular to it. A more detailed discussion of the triple differential cross sections as a function of the angles of the $\boldsymbol{k}_r$ and $\boldsymbol{k}_R$ motion with respect to the polarization axis as well as for the $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ motion will be the subject of a forthcoming paper.

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