01 Mar 2001

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Recommended Citation

R. Moshammer and B. Feuerstein and D. Fischer and A. Dorn and C. D. Schroter and J. Deipenwisch and J. R. Crespo Lopez-Urrutia and C. Hohr and P. B. Neumayer and J. H. Ullrich and H. Rottke and C. Trump and M. Wittmann and G. Korn and W. Sandner, "Non-Sequential Double Ionization of Ne in Intense Laser Pulses: A Coincidence Experiment," *Optics Express*, vol. 8, no. 7, pp. 358-367, Optical Society of American (OSA), Mar 2001.
The definitive version is available at https://doi.org/10.1364/OE.8.000358

Non-sequential double ionization of Ne in intense laser pulses: a coincidence experiment

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Abstract: The dynamics of Neon double ionization by 25 fs, 1.0 PW/cm^2 laser pulses at 795 nm has been studied in a many particle coincidence experiment. The momentum vectors of all ejected atomic fragments (electrons and ions) have been measured using combined electron and recoil-ion momentum spectroscopy. Electron emission spectra for double and single ionization will be discussed. In both processes the mean electron energies differ considerably and high energetic electrons with energies of more than 120 eV have been observed for double ionization. The experimental results are in qualitative agreement with the rescattering model.

OCIS codes: (020.4180) Multiphoton processes; (020.0020) Atomic and molecular physics

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1 Introduction

A detailed understanding of the processes occurring during the interaction of intense laser fields with matter form the basis for a wide range of potential applications from surgery and materials treatment to ignition of nuclear fusion. The cleanest way to address the most dominant process, i.e. ionization of bound electrons, is to study the response of single atoms exposed to intense ultrashort laser pulses. It is almost 20 years ago that surprisingly large yields for the creation of doubly charged ions in strong, linearly polarized laser pulses [1] were observed. While single ionization is well understood within the framework of the single active electron model [2], the yields for double ionization exceed the value one would expect assuming a sequence of individual and independent single ionization events during the laser pulse by many orders of magnitude. There is common agreement, that this non-sequential (NS) contribution to double ionization is a consequence of the electron-electron interaction but the underlying mechanism remained unclear over many years (for a review see [3]).

Essentially three mechanisms have been proposed to explain the enhanced ion yields. First, Fittinghoff et al. [4] proposed a "shake-off" mechanism as dominant contribution. Like in double ionization with single high energetic photons the sudden change of the screened atomic potential due to the escape of one electron causes ejection of the second electron. Second, and most recently, collective multi-electron tunneling has been considered but found to be quantitatively too weak [5]. A semi-empirical expression resembling a tunneling formula has been found that is in good overall agreement with measured multiple ionization rates. Third, a classical rescattering model was proposed by Corkum and Kulander et al. [6,7]. Within this model double ionization is considered as a two step process. First, one electron is removed from the atom by field ionization. Afterwards, this electron is driven by the oscillating electric field of the laser pulse and under certain conditions it is thrown back to its parent ion with sufficient kinetic energy to knock out a second electron in a (e,2e)-like collision. However, the rescattering mechanism for direct double ionization in He was found to be too unlikely to be the dominant process [8]. More refined time dependent quantum calculations [9] provided the first theoretical evidence in support of the rescattering

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mechanism. But, based only on high precision total ion yield measurements [10] as a function of laser intensity, a conclusive decision in favor of one of the proposed mechanisms for NS multiple ionization was not possible. Recently, the application of experimental imaging techniques, originally designed for the study of multi-electron processes in atomic collisions with charged particles and single photons [11], succeeded in measuring the ion momentum distributions for single, double and triple ionization of He, Ne and Ar [12,13,18]. The experimental findings for NS multiple ionization have been shown to be in agreement only with the rescattering model. Any mechanism based on a simultaneous release of two (or more) electrons at an instant when the electric field maximizes, like e.g. shake-off or collective tunneling, can be ruled out as a dominant contribution to NS multiple ionization. Those mechanisms as well as sequential multiple ionization should result in an ion momentum distribution along the laser polarization axis peaking at zero momentum. In contrast, the observed distributions exhibit distinct maxima at non-zero values. These results settled the long standing discussion about the mechanisms responsible for NS multiple ionization, and they initiated a series of theoretical investigations [14,15,16,17], which all include the recollision mechanism. As a continuation of these first differential experiments Weber et al. [18] were even able to obtain coincident two-electron momentum spectra for double ionization of Argon for both cases, sequential and non-sequential double ionization. In the regime of NS ionization they found a strongly correlated motion of electrons: In almost all cases both electrons are emitted into the same half-sphere. Because in this pioneering experiment only the momentum component along the light polarization axis could be measured, no information about e.g. the energy or angular distribution of ejected electrons was obtained. Charge state resolved electron energy distributions for single and double ionization of Xe have been measured recently by Witzel et al. [19]. They found slightly hotter electrons in coincidence with double ionization. To ultimately unravel the many-particle dynamics of double (multiple) ionization in intense laser fields would require a complete determination of the final state, i.e. the measurement of the vector momenta of all fragments (electrons and ions). Up to now, the accomplishment of such a kinematically complete experiment for double ionization was beyond experimental capabilities.

Here we report on a kinematically complete experiment on double ionization of Neon in ultra-short (25 fs FWHM) laser pulses at intensities (1.0 PW/cm^2) where NS double ionization dominates and single ionization proceeds via tunneling. The created ion and up to two electrons were detected in coincidence. Using Cold Target Recoil-Ion Momentum Spectroscopy (COLTRIMS) in combination with a highly efficient 4π electron spectrometer (so called reaction-microscopes [20]) the momentum vectors of all three particles and the charge state of the ion were determined.

2 Experiment

The experiment was performed at the Max-Born Institute in Berlin using a Kerr-lens mode locked Ti:Sapphire laser at 795 nm wavelength with pulse energy of 50 μJ at a repetition rate of 1 kHz. By focussing the 25 fs (FWHM) light pulses with a mirror (f = 100mm) to a spot of 8 μm (FWHM) diameter into an ultrahigh vacuum chamber ($2 \cdot 10^{-11}$ torr), a pulse peak intensity of 1.0 PW/cm^2 was reached. Intensity fluctuations were monitored during the experiment and kept below 5%.

At its focus the laser beam was crossed by a low-density (10^8 $atoms/cm^3$) supersonic neon jet formed by expanding Ne gas at a pressure of 5 bar through a cooled (LN_2 temperature) 10 μm nozzle. After expansion, the beam was collimated over a total length of 2 m to a rectangular shape of 0.05×2 mm^2 at the interaction point, oriented with the broad side perpendicular to the laser beam. This resulted in a an overlap volume of laser beam and neon jet of 50μm length and 8 μm diameter.

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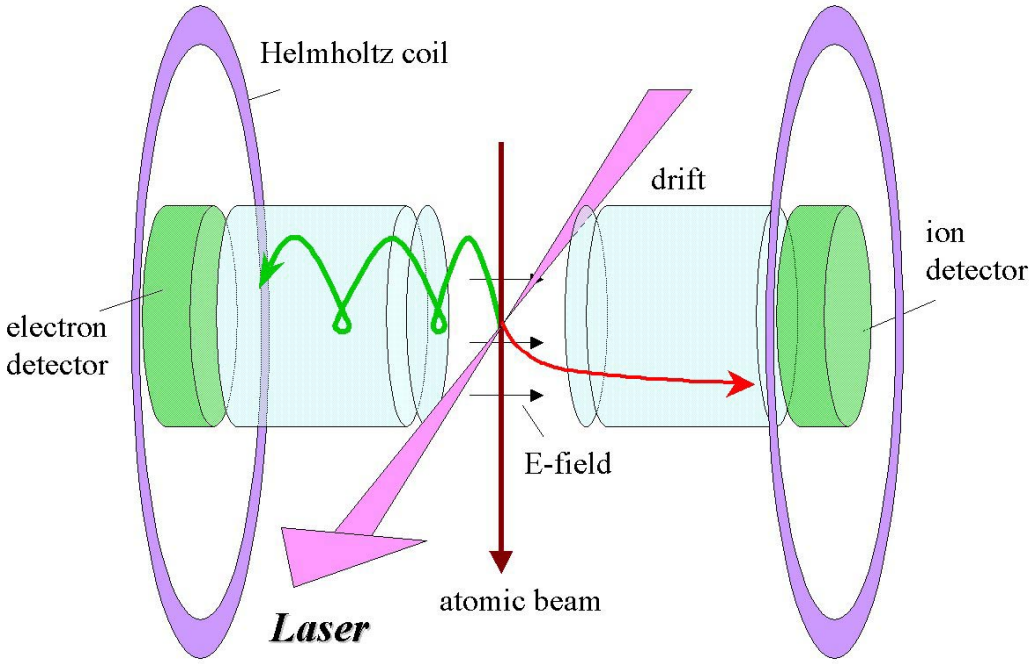

Fig. 1. Schematic drawing of the experimental setup.

Electrons and ions created in the overlap volume were extracted into opposite directions by a weak 1 V/cm electric field acting over 22 cm perpendicular to the laser beam and parallel to the light polarization direction (see Fig. 1). An additional homogeneous magnetic field of 12 Gauß ($\Delta B/B < 10^{-3}$) generated by two Helmholtz coils forced electrons with nonzero transverse energies to spiral trajectories. In this way all electrons with transverse energies below 60 eV and all ions were projected onto large area (75 mm diameter) position sensitive detectors placed 30 cm away from the reaction zone. The electron detector was multi-hit capable enabling the registration of position and time of two electrons emerging from the same atom if they arrived at the detector with a spacing in time of more than 15 ns. The ion charge state and the momentum vectors of both, ions and electrons, were calculated from their measured absolute flight times and their positions on the detectors. In order to ensure that electron-ion pairs registered in coincidence emerged from the same atom, the target and the residual gas densities had to be such that less than one ionization event per laser pulse occurred. This resulted in almost background free time of flight spectra (Fig. 2) at ion count rates between 10 and 100 Hz. The effective coincidence count rate for true double ionization events was only about 1 per minute. In order to collect a statistically significant amount of double ionization events a beam-time of several days was necessary putting high demands on the long term stability of the laser system.

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Received February 05, 2001; Revised March 01, 2001

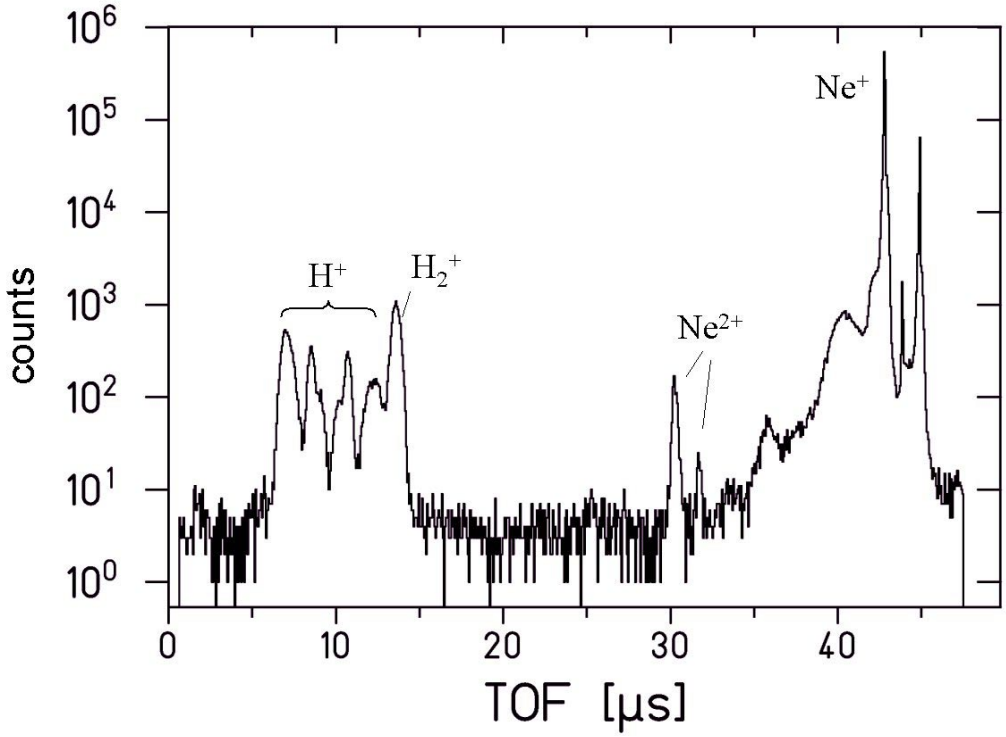

Fig. 2. Ion time-of-flight spectrum at a laser intensity of 1.0 PW/cm^2. The different Ne^{1+} peaks reflect the Ne isotope abundance. The H-ions are residual gas contributions.

If one assumes that electrons and ions experience the light field for the same time (short laser pulses) and that only a very small momentum is transferred to the atom by the absorbed photons then the ion momentum balances the sum momentum of the ejected electrons. Thus, the detection of only two out of three fragments created in a double ionization event would be sufficient to determine the momentum of the missing third particle. This assumption as well as the correct assignment of coincident registered ions and electrons can be checked for the case of single ionization by plotting the Ne^{1+} momentum component along the light polarization axis (longitudinal momentum) versus that of the electron (Fig. 3). The distinct accumulation on the diagonal line indicates that the momenta of ions and electrons are equal and opposite in sign for each single event. This proves, that the aforementioned assumption of negligibly small net momentum transfer from the laser field is valid and that false coincidences, in which the detected ion and electron are created in the same laser pulse but do not come from the same atom, are unlikely. We therefore measured the Ne^{2+}-ion in coincidence with one electron and deduced the vector momentum of the second electron using momentum conservation. This way the vector momenta of all ejecta have been determined giving access to the complete kinematics of NS double ionization in strong laser fields. The longitudinal momentum resolution is controlled experimentally (Fig. 4) and a value of $(p_{e\|}^2 + p_{ion\|}^2)^{1/2} = \pm 0.055$ a.u. has been achieved for single ionization. Thus, the recoil-ion spectrometer is sensitive to changes in ion energies on the sub-µeV level. It is this combined resolution that also mainly defines the error in the determination of the second electron in the case of double ionization.

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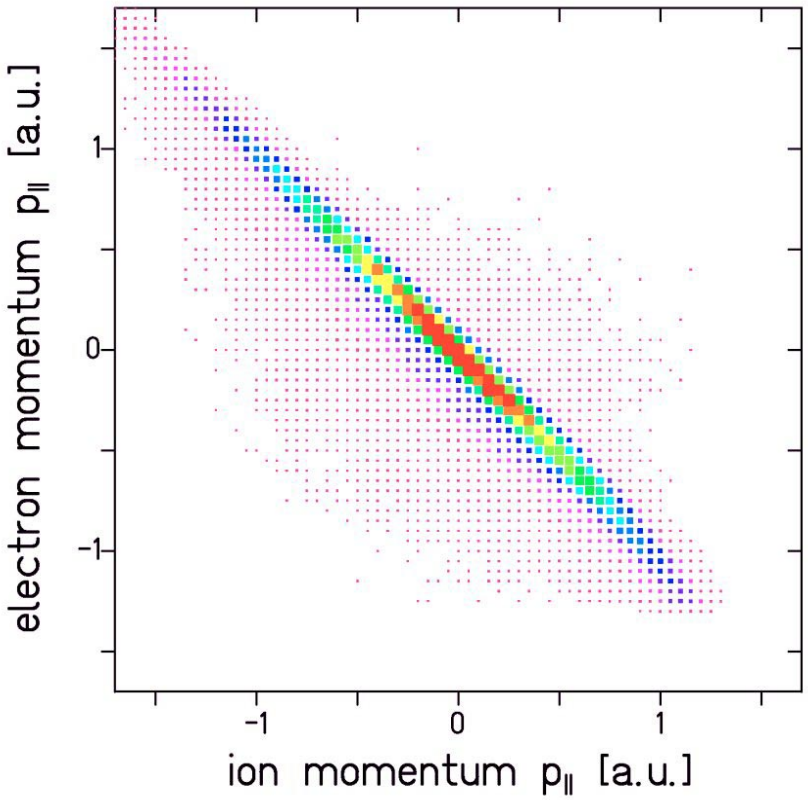

Fig. 3. Momentum correlation between Ne^{1+} ions and electrons which were registered in coincidence. Plotted are the ion and electron momentum components along the polarization of the laser field at a peak intensity of 1.0 PW/cm^2. (log. Scale).

In the direction perpendicular to the polarization axis along the jet expansion the ion momentum resolution is limited by the internal jet temperature to 0.2 a.u.. The transverse electron momentum resolution depends on their flight times. For those equal to an integer number of the cyclotron revolution time all electrons are focused on the center of the electron detector independent of their transverse energy. The resolution is best ($\Delta p_{e\perp}$ = 0.05 a.u.) for those electrons, which arrive with flight times exactly between two focal points. Averaged over all possible flight times a mean value for the transverse momentum resolution of better than $\Delta p_{e\perp}$ = 0.1 a.u. is estimated.

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Received February 05, 2001; Revised March 01, 2001

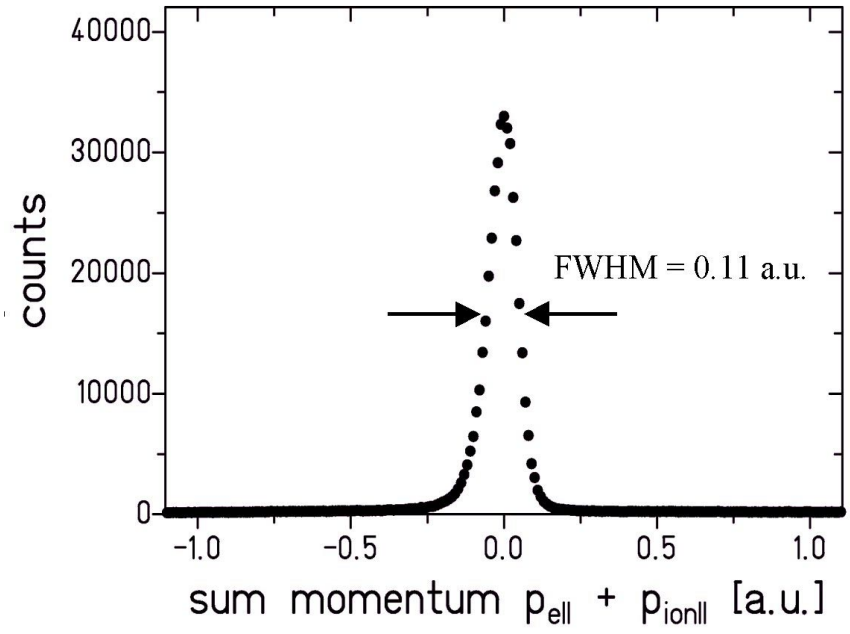

Fig. 4. Longitudinal sum momentum of electrons and Ne^{1+} ions.

3 Results

In the experiment both processes, single and double ionization, are measured simultaneously yielding electron emission spectra for defined ion charge states. Furthermore, since for each ionization event the electron vector momenta are recorded irrespective of the emission angle for energies up to 60 eV the whole electron emission distribution is measured at once. The obtained electron energy as a function of the emission angle with respect to the light polarization axis is shown in Fig. 5 for single ionization of Ne. At the present laser intensity of 10^{15} W/cm^2 single ionization proceeds via tunnel-ionization. After being released the electron performs a quiver motion in the external laser $E(t) = E_o(t) \cdot \sin(\omega t)$ and the finally transferred drift momentum $p_{drift} = \omega^{-1} \cdot E_o(t_1) \cdot \cos(\omega t_1) = 2 \cdot U_P^{1/2} \cdot \cos(\omega t_1)$ after the end of the laser pulse depends on the phase ωt_1 where ionization occurred. (U_p = 60 eV is the ponderomotive potential at 10^{15} W/cm^2.) This drift momentum is small if the electron is set free at a phase where tunnel ionization occurs most likely, namely when the electric field is close its maximum. Because the drift momentum, though small, pushes electrons along the light polarization axis, the angular distribution peaks at $\vartheta = 0$. Hence, single ionization is expected to be dominated by electrons with low energies and small emission angles in agreement with the experimental data and tunnel ionization theory [2](see also [13]).

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Received February 05, 2001; Revised March 01, 2001

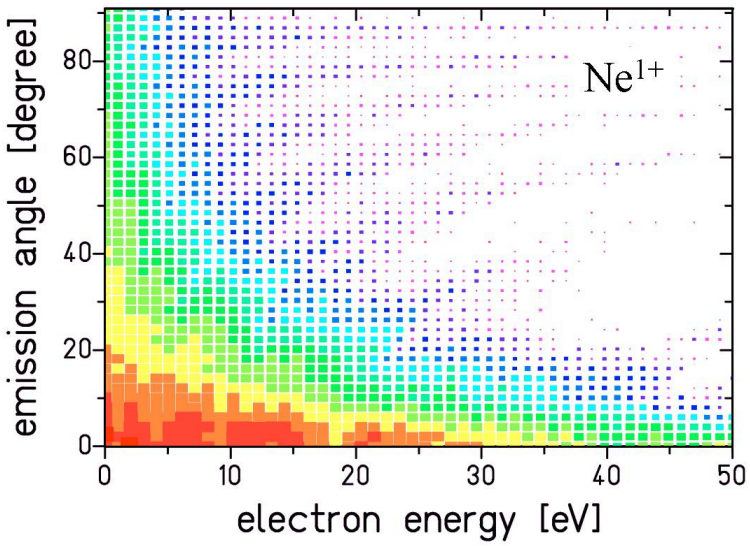

Fig. 5. Electron energy versus emission angle for Ne single ionization. $\vartheta = 0$ corresponds to emission along the light polarization axis. (log. Scale).

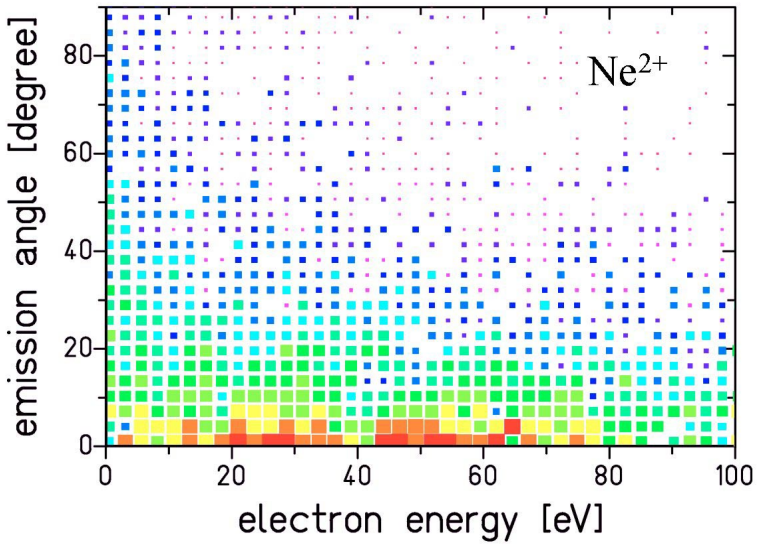

Fig. 6. Same as Fig. 5 for double ionization. Notice the different energy scale.

In contrast to the single ionization results high energetic electrons with energies up to more than 120 eV have been observed for double ionization (Fig. 6). The emission characteristics of one typical electron is displayed irrespective of the energy and angle of the other electron or the Ne^{2+} ion. We will briefly discuss how this finding can be explained with the classical rescattering model. Within this model doubly charged ions are most efficiently created according to the following scenario. The first electron is tunnel ionized at a phase close to 107 degree. After an excursion of almost 200 a.u. away from the core this electron is thrown back to its parent ion with a kinetic energy of $3.17 \cdot U_p$, which is sufficiently high to overcome the binding energy I_p and to ionize a second electron in an inelastic encounter. This (e,2e) collision occurs at a phase when the electric field is close to a zero crossing. We will consider

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this phase as a time mark where both electrons and the Ne^{2+} ion are born. Afterwards, each electron will gain an additional drift momentum of almost $p_{drift}^{max} = 2 \cdot U_P^{1/2}$ in the direction of the electric field vector. Hence, if an electron is born with zero kinetic energy it appears after the laser pulse with an energy of $2 \cdot U_p$ = 120 eV and it will be emitted in the direction of the polarization axis. The electron energy distribution for small emission angles (projection from Fig. 6 for $\vartheta = 5 \pm 5^o$) is shown in Fig. 7a. The distribution exhibits a clear edge close to $2 \cdot U_p$ (arrow in Fig. 7a). Electrons emitted into the transverse direction (Fig. 7b; $\vartheta = 80 \pm 10^o$) have very low energies. In general, though, the electrons are not born with vanishing kinetic energy because the excess energy ($3.17 \cdot U_P - I_P$) of the recolliding projectile electron is shared between the two electrons. It can be shown easily, that a forward scattering of the projectile electron leads to final electron energies below $2 \cdot U_p$ and that higher energies can be attributed to backward scattering [21]. From field free (e,2e) collisions it is well known that small angle forward scattering is by far more likely. Thus, it is not surprising that most electrons appear with energies between zero and $2 \cdot U_p$. Interestingly, elastic backward scattering of the returning electron has been identified [22] in single ionization as a plateau in the electron energy distribution which extends up to $10 \cdot U_p$.

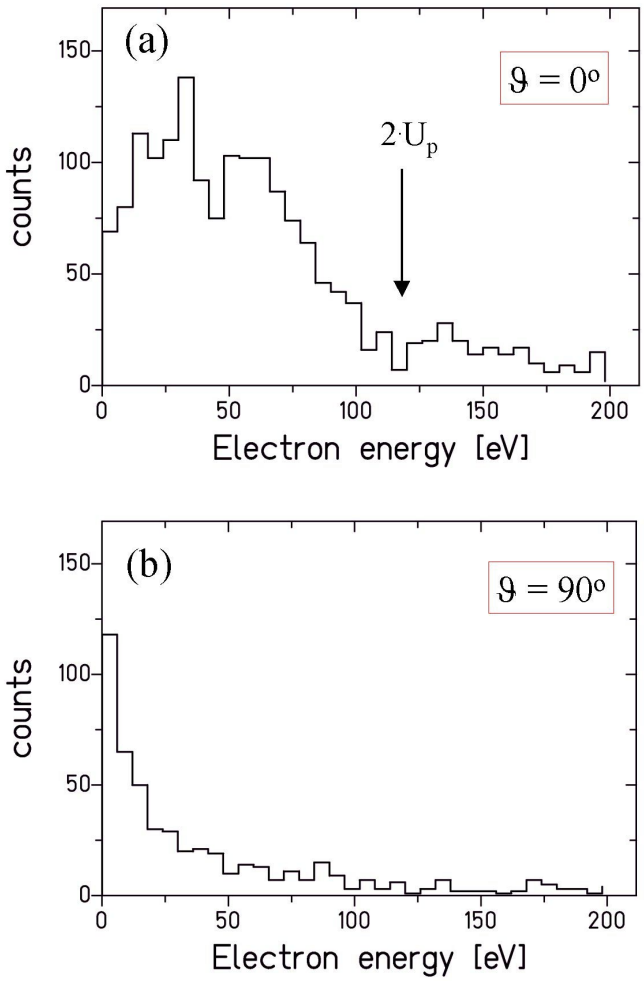

Fig. 7. Electron energy distributions for emission along (a) and perpendicular (b) to the polarization axis in double ionization.

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In conclusion, the presented experimental results are in qualitative agreement with the rescattering model. Even though the electron motion is dominated by the ponderomotive force the signature of the (e,2e) collision dynamics is still preserved in the final state. In this paper only a very small subset of the collected experimental data could be discussed. In particular the evaluation of the momentum balance in the transverse direction, which is not influenced by the ponderomotive force, as well as the analysis of correlated two electron spectra will unravel more details of the underlying many-particle dynamics in strong laser fields. These data will be discussed in a subsequent publication.

In the future, experimental studies of electron correlation in strong laser fields are in the realm of possibility, and a large variety of targets, like e.g. atoms, molecules and clusters can be investigated using advanced many particle coincidence techniques. On this road in particular the performance of a kinematically complete experiment on Helium, the most simple two-electron system, remains as an experimental challenge.

Acknowledgments

This work has been supported by the Deutsche Forschungsgemeinschaft (DFG, number RO 1029/2-1) and the Leibniz-Program of DFG. Support from GSI is gratefully acknowledged. The Freiburg group would like to thank for the hospitality of the MBI. We are grateful to R. Dörner and W. Becker for numerous discussions.

#30944 - $15.00 US Received February 05, 2001; Revised March 01, 2001