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Correlated Electron Emission in Multiphoton Double Ionization

Th. Weber[1], H. Giessen[2], M. Weckenbrock[1], G. Urbasch[2], A. Staudte[1], L. Spielberger[1], O. Jagutzki[1], V. Mergel[1], M. Vollmer[2], and R. Dörner[1*]

[1] Institut für Kernphysik, Universität Frankfurt,
August Euler Str. 6
D-60486 Frankfurt, Germany
[2] Fachbereich Physik, Philipps-Universität,
Renthof 5, D-35032 Marburg Germany
* to whom correspondence should be addressed.
E-mail: doerner@hsb.uni-frankfurt.de

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Electronic correlation governs and fuels the dynamics of many important phenomena in nature, from chemical reactions to solid state effects (e.g., superconductivity). The nature of such correlation effects can be most clearly addressed in single atoms. Therefore the emission of two electrons from an atom induced by impact of a single photon [1], a charged particle (see e.g. [2]) or in a short laser pulse [3] have become the paradigmatic processes for the study of dynamical electron correlation in physics. For atoms and molecules exposed to laser fields that are comparable with the nuclear fields extremely high probabilities for double ionisation have been found [4, 5] and attributed to electron-electron interaction [3]. Here we report the observation of strong correlation of the magnitude and the direction of the momentum of two electrons that are emitted from an argon atom, driven by a femtosecond laser pulse (at 38 TW/cm^2). Upon increasing laser intensity, however, this momentum correlation between the electrons is lost, demonstrating a transition in the laser-atom coupling mechanism.

The interaction of superstrong laser fields with matter has a wide range of applications, from surgery to ignition of nuclear fusion. The underlying fundamental physical processes of this interaction, however, still bear many open questions. An extremely clean way to address them is to place just one single atom in the laser focus and examine its reaction. In many cases the response of the atom to the strong laser field can be very well understood by considering only one of its electrons as active. One important experimental observation, however, withstands explanation in such simple terms: the production of doubly and multiply charged ions is by many orders of magnitude more effective than predicted by an independent-electron model [4, 5]. Since this discovery it was agreed upon that we need models that include electron-electron correlation to explain this enhanced multiple ionization rate. Intense efforts to model the two-electron process of double ionization in a laser field have recently reproduced the main feature of a knee structure in the double ionization yield as function of laser peak intensity [6, 7, 8, 9, 10, 11] and, in some cases, moreover yielded quantitative agreement with the experiment [12, 13].

The physical mechanism behind this nonsequential process, however, is still debatable. The most widely accepted scenario is a rescattering mecanism [14, 6] in which the first electron is accelerated by the laser field and later driven back by the field towards its parent ion. The second electron is then liberated in an electron-ion impact ionization (see, e.g. [6, 15, 8, 9, 10] for arguments supporting this view and [16, 7] opposing this model). Alternatively, Fittinghoff and coworkers [4] have suggested that the second electron is shaken off via the same mechanism as in double ionization by a single high energy photon (e.g., from synchrotron radiation). Recently, Becker and Faisal have suggested a model of an energy sharing mechanism using coulomb correlation represented by a single Feynman diagram [12, 17]. Strong experimental evidence supporting the rescattering/energy sharing mechanism is provided by the supression of double ionisation in elliptically polarized laser fields [15].

In this paper we report the direct experimental observation of strong electron-electron correlation in the final state. We show that the momentum of one electron strongly depends on the momentum of the other electron. Such coincident electron momentum distributions serve as a detailed test ground for the rivaling theoretical approaches. But they also yield direct and intuitive insight into the dynamics of the laser-atom interaction and shade new light on the mechanism leading to double ionization. They answer the question whether the two electrons are pulled by the the laser field to the same side or whether the electron-electron repulsion pushes them to opposite sides. The latter is observed with synchrotron radiation at high photon energies, where the energy of just one photon is sufficient to liberate both electrons [1].

A very first step towards differential information on the correlated multiple ionization in strong fields has been gained only recently by observing the momentum distribution of the doubly charged ions for helium [18, 19] and neon targets [20]. Both studies found large ion momenta which are not peaked at zero, highlighting the correlated nature of the process. In contrast, single ionization and thus any uncorrelated process [19] gives maximum ion yield at zero momentum.

We have used the well established technique of COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) (see [21, 22] for recent reviews) for the experiment. The linearly polarized light from a Ti-sapphire laser (800 nm, 220 femtoseconds pulse width) was focussed by a 5 cm focal length lens onto a precooled supersonic Argon gas jet.

The ions and electrons are guided by an electric field to opposite sides towards two position sensitive channel plate detectors with delay-line readout. Their momenta are determined from the flight times and positions of impact. For more details of the experimental setup and data analysis see [23]. The polarization axis of the light was parallel to the electric field and perpendicular to the direction of the gas jet. The peak laser intensity was determined by ion yield measurements to an estimated accuracy of 15%.

The local Argon pressure in the gas jet was adjusted such that the average ion count rate was about 0.04-0.08 per shot. The simultaneously measured coincidence between Ar^{1+} ions and electrons allowed an online monitoring of the fraction of false coincidences, in which the detected ion and electron are produced in the same laser shot but do not come from the same atom. Such events (about 30% of the total counts) are localized outside the diagonal line in figure 1. The contribution of false coincidences has be subtracted from the Ar^{2+}-electron coincidence data. This background correction, however, changes only the details, but not the major findings of our results.

The localization of the correlated events on the diagonal line demonstrates, that the momenta of Ar^{1+} ion and electron are equal and have opposite sign for each single event. This proves that the laser pulse is short enough to prevent the electron from leaving the focus during the pulse. Therefore, the momentum transfer from the optical field to the electron and Ar^{1+} ion is equal and opposite, both at every individual time interval and hence for the whole pulse duration. The laser does not pass on significant net momentum to the electron-ion system. The photon momentum is negligible on the scale of interest here, as 1.5 eV photons have a momentum of only 0.0004 a.u. (atomic units: 1 a.u. of momentum is equivalent to $1.995{\cdot}10^{-24}$ kg m/s). This is essential for the present experiment, since it assures that for double ionization the ion momentum k_{ion} is given by the sum of the two electron momentum vectors $k_{e1,2}$, $k_{ion} = -(k_{e1} + k_{e2})$. We have therefore measured k_{ion} and the momentum of one of the electrons and can then deduct the momentum of the second electron from momentum conservation. The horizontal line-structure that is visible in figure 1 results from above-threshold ionization peaks (see e.g., [24]). These peaks are resolved for slow electrons whereas they are neither resolved in the ion momenta nor for faster electrons, since we use a time-of-flight technique. The overall resolution (the width of the diagonal of momentum conservation) is about 0.4 a.u. FWHM.

The main result of our work is shown in figure 2. The vertical axis shows the momentum component of the measured electron in the direction of polarization (k_{ez1}) and the horizontal axis the same momentum component of the second electron. The data are integrated over the transverse momentum of the first and second electron. The momentum of the second electron is deduced from the measured momenta of the first electron and the recoiling ion by momentum conservation.

At $3.8{\cdot}10^{14}\mathrm{W/cm}^2$, right at the knee of the double ionization rate (see [25, 19]), a strong momentum correlation between the two electrons is found. There is a clear maximum for both electrons being emitted with the same momentum component of about 1 a.u., whereas emission to opposite half planes is strongly suppressed. At an intensity of $15{\cdot}10^{14}\mathrm{W/cm}^2$ (figure 2b), in the regime where the ratio of double to single ionization steeply rises with laser peak power, the correlation between the electrons is completely lost. Since only one component of the electron momenta is shown, figure 2a does not imply that the electrons are really emitted in parallel. More likely, there will be some angle between the electrons due to the repulsion.

The difference between figure 2a and 2b, which is accompanied by a factor of six increase in the ratio of double to single ionization [25, 19], is a direct experimental proof for the change of mechanisms for double ionization as a function of laser power. In the regime of the knee in the double ionization rate, the two-electron process is mediated by electron-electron correlation, whereas at high laser powers, double ionization proceeds by sequential emission of the two electrons. Since in this case there may be a random number of optical halfcycles between the emission of the two electrons, no directional correlation between the electrons can be expected.

An instructive alternative perspective on figure 2 is obtained by rotating the distribution by 45 degrees. The diagonal coordinate frame as indicated in the figure shows the sum and difference momenta $k^+ = k_1 + k_2$ and $k^- = k_1 - k_2$ of the individual electron momenta $k_{1,2}$. Due to momentum conservation, k^+ is equal and opposite to the Ar^{2+} recoil-ion momentum (compare [18, 19, 20]). Figure 2a shows that the width of the k^- distribution is narrower than the width in the k^+ direction. The coordinates k^+ and k^- are helpful to illustrate the relative importance of the two counteracting effects of electron-electron repulsion and acceleration of particles by the optical field. Both influence the observed final state momenta in different ways. Electron repulsion (and two-body electron-electron scattering) does not change k^+ but contributes to the momentum k^-. On the other hand, once both electrons are set free, the momentum transfer received from the field is identical. Therefore this part of the acceleration does not change k^-

but adds to k^+. The observed wide k^+ and narrow k^- distributions thus indicate that the joint acceleration of the electrons in the laser field clearly dominates over the influence of electron repulsion. *Both electrons are driven by the laser field to the same side.* This experimental finding is in agreement with recent theoretical results calculated within the model of the energy sharing mechanism [26].

How does this finding relate to the two aforementioned correlation mechanisms of rescattering and shake-off? The pure case for the latter is found in single-photon double-ionization, where the final state momentum distribution is predominantly determined by the long-range Coulomb interaction in the final state. It forces the electrons almost completely to opposite half spheres (see [1] for a recent review). At small energies, which dominate in our case, k^+ is much smaller than k^- (see [27]). If a shake-off process were to take place in a strong laser field, it would set both electrons free in a time interval very short compared to the optical cycle. Furthermore, shake-off would have its highest probability at the maximum intensity of the electric field, where also single ionization maximizes. After beeing set free the electrons perform a quiver motion in the field. The remaining net momentum transfer at the end of the laser pulse depends on the phase at which the electrons are set free. Ionisation at the maximum of the field corresponds to zero net momentum transfer from the field. Therefore, the field is not expected to play a significant role for the final state electron momenta resulting from a shake-off. Such electrons should be found either close to the origin in figure 2 or in the region of opposite momenta due to electron repulsion. *Thus figure 2a is in clear contradiction to the expectations of a shake-off model.*

In the rescattering or 'energy-sharing model', there is one particular process which produces electrons with similar momentum. If the energy of the primary electron at its reencounter with the ion is equal to the energy needed to excite the second electron in the ion, the excitation process would stop the primary electron. The excited electron will then be set free with very little excess energy by the laser. Both electrons will finally have similar momentum which is only determined by the phase of the field at the reencounter. The momentum of about 1 a.u. corresponds to a phase at rescattering of 35 deg and a return energy of the primary electron of about 18 eV. This is sufficient for an excitation, since a broad band of many excited states of the Ar^{1+} ion starts at about 16.5 eV (the ionization energy is 27 eV). We conclude that the rescattering model provides a plausible scenario to produce the observed momentum correlation of the electrons.

Our measured momentum correlation shows that it is not the electron correlation in the initial ground state of the atom which leads to the enhanced double ionisation in strong fields, but rather a rescattering which is a result of the driving field. The magnitude of the momenta of the electrons in the direction of the laser field provide information on the phase of the field at the instant of recollision. The width of the distribution in the k^- direction is sensitive to the details of the rescattering. Thus we expect that the momentum in the k^- direction and the momentum correlation in the directions perpendicular to the polarization will give further insight into the details of the correlation mechanism in future studies.

Finally, we note that the observed emission of the electrons to the same side confirms a recent prediction of Taylor and coworkers [28]. They have solved the time-dependent Schrödinger equation for two electrons in an optical field in three dimensions and found that most of the double ionization probability flux emerges to the same side. Similar conclusions have been drawn from one-dimensional model calculations [29].

The findings of this work provide a benchmark for theoretical efforts to calculate one of the most simple correlated two electron processes in nature. Our demonstrated method using many-particle momentum space imaging for strong field physics opens up the road for a variety of applications. The study of electron correlation in strong fields is now possible in molecules, clusters or solids (such as superconductors).

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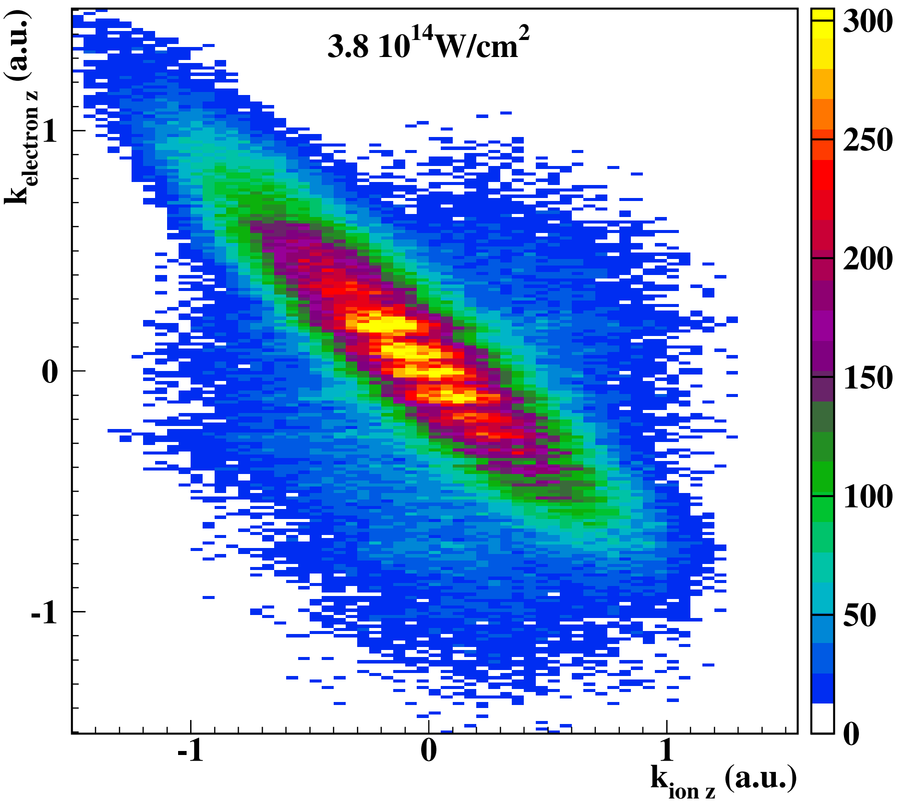

Figure 1: Momentum correlation between Ar^{1+} ion and electrons created in the focus of a 220 fsec, 800nm laser pulse at peak a intensities of $3.8\cdot 10^{14}$ W/cm^2. The horizontal axis shows the momentum component of the Ar^{1+} ion along the polarization of the laser field, the horizontal axis the momentum of the detected electron.

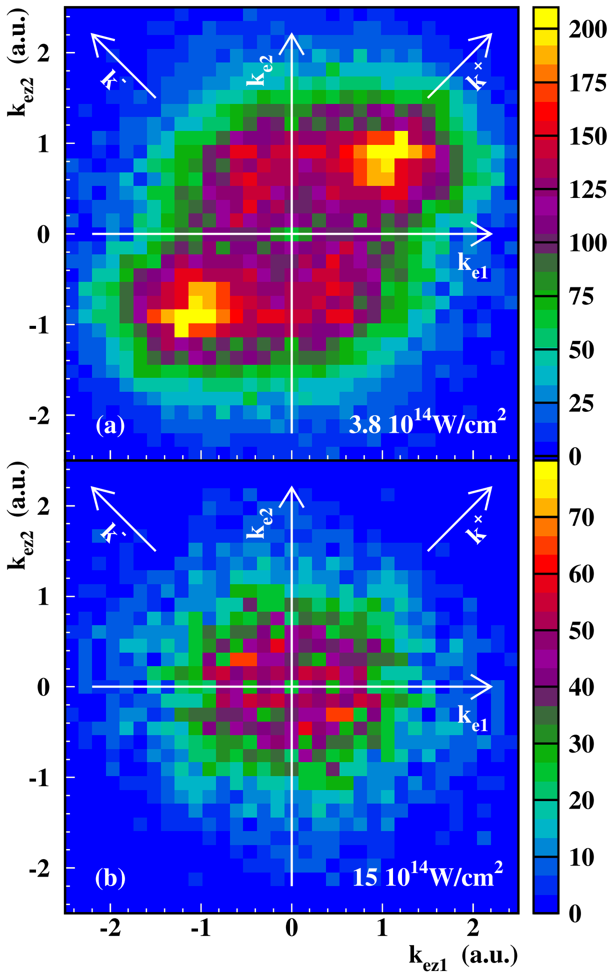

Figure 2: Momentum correlation between the two emitted electrons when a Ar^{2+} ion is produced in the focus of a 220 fsec, 800nm laser pulse at peak intensities of $3.8\cdot 10^{14}$ W/cm^2 (a) and $15\cdot 10^{14}$ W/cm^2 (b). The horizontal axis shows the momentum components of one electron along the polarization of the laser field, the vertical axis the same momentum component of the corresponding second electron. Same sign of the momenta for both electrons means emission to the same half sphere. The data are integrated over the momentum components in the direction perpendicular to the polarization. The color coding shows the differential rate in arbitrary units.