J. Phys. B: At. Mol. Opt. Phys. **34** (2001) L449–L455 www.iop.org/Journals/jb PII: S0953-4075(01)25133-5

LETTER TO THE EDITOR

Experimental evidence for electron repulsion in multiphoton double ionization

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Received 22 May 2001, in final form 13 June 2001

Abstract
We have measured the coincident momenta of both electrons emitted in argon double ionization at 780 nm and 4.7×10^{14} W cm^{-2}. For electrons which are emitted with very small momentum transverse to the electric field, we find that the two electrons have highly unequal momentum. This is in contrast to the situation for larger transverse momenta for which most electrons are found with similar momentum. We interpret this observation as a manifestation of electron repulsion. We also identify contributions from electron impact excitation.

Strong field double ionization has been studied heavily since the advent of powerful short-pulse laser sources (see [1] for a recent review). From the observed rates for double ionization it became clear that in an intermediate regime of laser power double escape is mediated by electron correlation [2–5]. This has been confirmed by numerous theoretical calculations (see e.g. [6–14]). The most likely scenario today for the correlation dynamics is given by the rescattering model [15], which also successfully describes higher harmonic generation and the creation of high-energy electrons. For double ionization to occur in this model, one electron is set free, for example, by tunnel ionization, is accelerated and then driven back by the field to its parent ion. Upon recollision the ion is excited or ionized. The gross features observed in recent differential measurements of the doubly charged ion momenta [16–18] and electron energies [19, 20] could be successfully interpreted within this simple model [21] (for elaborate calculations reproducing the ion momentum distributions see [22–27]).

The first experimental work observing the correlated momentum of the electrons [28] showed a collective emission of both electrons to the same half sphere. It is most likely that the electrons were found to have the same momentum component along the polarization axis (parallel momentum $p_{\|}$). At first sight, this is surprising since electron repulsion would push the electrons in opposite directions. The great importance of such final state electron repulsion becomes evident in single-photon double ionization, which has been studied in great detail using synchrotron radiation. (See [29] for a recent review and [30] for a comparison of the findings for single- and multiphoton double ionization.) In the strong field case the electron and ion final state momenta are influenced by both electron repulsion and the action of the laser field on the charged particles. While the effect of the acceleration in the laser field is obvious in the coincident electron momenta [28], surprisingly the influence of electron repulsion is not evident in any of the previous experiments.

In the present experiment we succeeded in measuring not only one component of the electron momentum as in [28] but also the full momentum vector. We show that for such a more complete experiment the effect of the repulsion becomes visible.

The experiment was performed using COLTRIMS (cold target recoil ion momentum spectroscopy) (see [31] for a recent review). An animated figure of the experimental set-up can be found in [30]. In brief, the laser pulse (150 fs, 780 nm) has been focused by a lens of 5 cm focal length into a supersonic argon gas jet. The ions were guided by a weak electric field (3.5 V cm^{-1}) onto a position-sensitive channel plate detector with delay-line readout [32]. From the position of impact and the time-of-flight the three-dimensional momentum vector can be determined. The electrons are guided by the electric field and a parallel magnetic field (10.4 G) onto a second position-sensitive detector. The electron momentum vector is determined from the time-of-flight and the position of impact (see [33] for more detail on such magnetically confined electron imaging). The target density was adjusted to yield an ion rate of about 0.12 per shot. A remaining contribution from random coincidences, i.e. events where two or more atoms have been ionized in one shot, has been subtracted from the data. Qualitatively this background subtraction does not affect the momentum distributions presented here. From the measured momenta of one electron and the doubly charged ion the momentum of the second electron was deduced using momentum conservation. The transverse momenta of the ions are of the order of the experimental resolution of our system, thus no meaningful transverse momenta could be obtained for the second electron. For the detected electron our momentum resolution was about 10^{-3} au in the parallel direction and 10^{-2} au in the perpendicular direction. The absolute laser peak power was determined by fitting our relative measured Ar^{1+} rates to the calculated dependence of the rate on the laser peak power. In order to do this we used S-matrix rate calculations introduced by Becker and Faisal to calculate the pulse length and focal condition utilized in our experiment.

Figure 1 presents the main result of our work. The vertical axis shows the momentum component of the electron in the direction of polarization (p_{ez1}) and the horizontal axis the same momentum component of the other electron. For one of the electrons the transverse momentum was fixed to a certain interval. We observe a strong dependence of the correlation pattern on this transverse momentum. If one electron is emitted off the polarization axis, then both electrons are most likely found to have similar parallel momenta. In contrast, if one electron is fixed along the polarization axis with very small transverse momentum (figure 1(*a*)), then we usually find one fast and one slow electron.

We interpret our finding as a direct consequence of electron–electron repulsion. The $1/r_{12}$ potential forces the electrons into different regions in the three-dimensional phase space. As a consequence, for electrons to have equal parallel momentum requires some angle (i.e. transverse momentum) between them. The peak at $p_{ez1} = p_{ez2} = 1$ au is therefore most pronounced if at least one of the electrons has considerable transverse momentum. If in contrast one electron is fixed to move along the polarization axis the second electron is repelled longitudinally to be either faster or slower than the first one.

To discuss the measured distribution more quantitatively figure 3 shows the classically allowed region of phase space calculated within the rescattering model. We discuss the figure using the classical phase relations shown in figure 2. If the primary electron tunnels at a phase between 0° and 90° the electron will not return, between 90° and 91.5° it will return with an

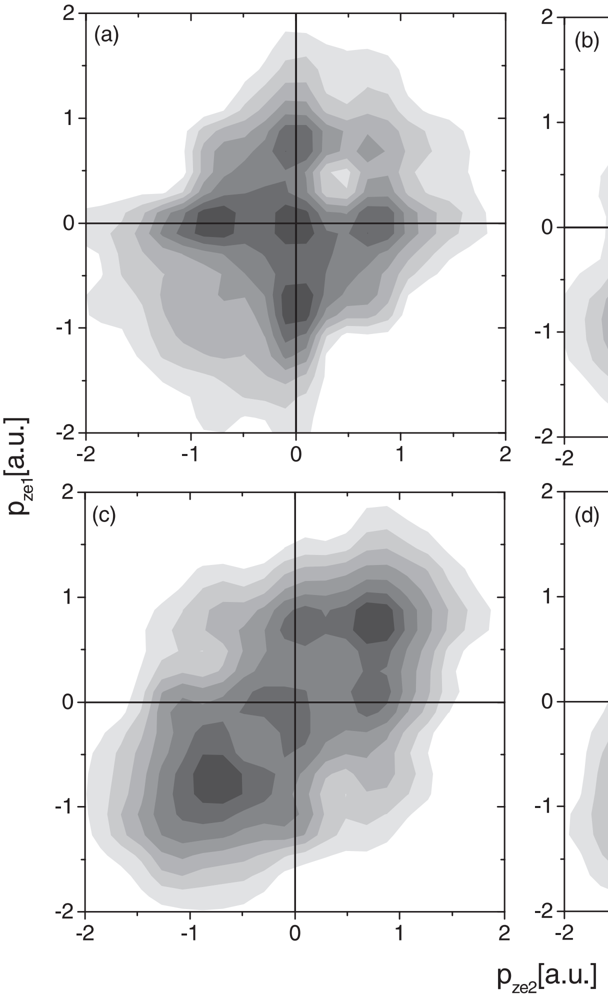

Figure 1. Momentum correlation between the two emitted electrons when an Ar^{2+} ion is produced in the focus of a 150 fs, 780 nm laser pulse at peak intensities of 4.7×10^{14} W cm^{-2}. The horizontal axis shows the momentum components of one electron along the polarization (p_{ez1}) of the laser field and the vertical axis the same momentum component of the corresponding second electron. The same sign of the momenta for both electrons means emission to the same half sphere. (*a*) One of the electrons has a transverse momentum $p_\perp < 0.1$ au; (*b*) $0.1 < p_\perp < 0.2$ au; (*c*) $0.2 < p_\perp < 0.3$ au; (*d*) $0.3 < p_\perp < 0.4$ au. The colour coding shows the differential rate in arbitrary units.

energy below 15.8 eV, insufficient to excite or ionize the Ar^{1+} ion (see figure 2) hence not leading to double escape. Between 91.5° and 93° the initial phase corresponding to a return phase between 405° and 418° (figure 2(*b*)) the recollision energy is above the first excitation threshold (15.755 eV) and below the ionization threshold of Ar^{1+} (27.6 eV) (region between i and ii in figure 2). In this case the ion can be excited and the primary electron loses the according amount of momentum. Since the field at the return or very shortly after is so high that most of the excited states are already over the barrier (figure 2(*c*)) the excited electron will be liberated instantaneously. From then on both electrons will be accelerated in phase, one starting with zero momentum the other with the left-over energy from the excitation process. This yields a

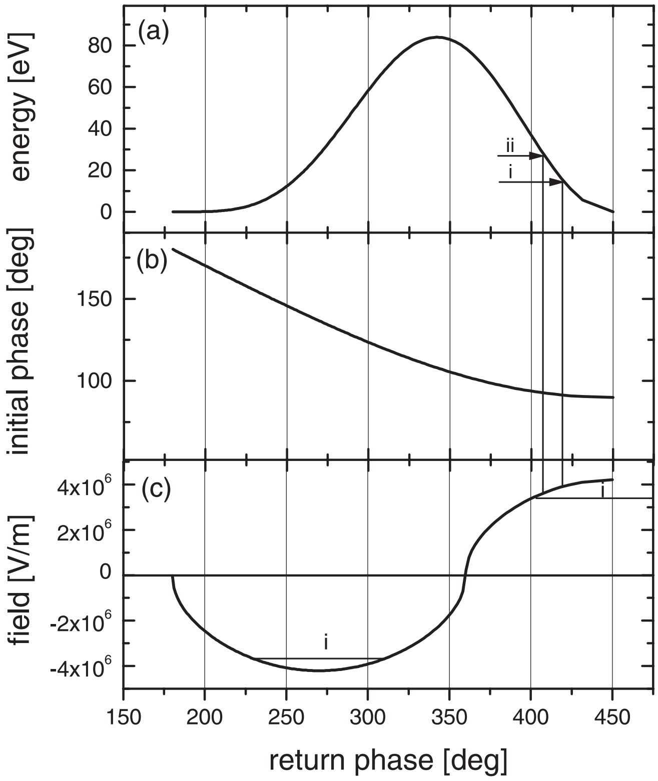

Figure 2. Phase relationships within the rescattering model for 780 nm at intensity 4.7×10^{14} W cm^{-2}. The horizontal axis gives the phase of the laser field at the instant of recollision: (*a*) return energy of the rescattered electron; (*b*) initial phase at which the electrons were set free. The arrows i and ii indicate the threshold for excitation and ionization, respectively; (*c*) electric field at return. The curves indicate the critical field for above the barrier ionization of the first excited states i of Ar^{1+}. In most cases excitation occurs at return phases where the field is high enough to quench the excited state instantaneously.

fix relation between the momentum of the primary electron and the second electron for each given excitation energy. This band of phase space for recollision with excitation is shown by the medium-grey zig-zag line. The different points along this line will be populated with extremely different probability. This probability will be strongly influenced by the product of the tunnelling probability at the respective starting phase of the primary electron times the excitation cross section. This will result in a strong maximum at the end of the line indicated by the black area. As already discussed in [28] the location of the peak of the intensity is close to this point in phase space. Therefore we believe that rescattering with excitation is likely to be responsible for this main peak. This conclusion from our data is further supported by recent rate calculations showing a rise by a factor of 2 in the rate of doubly charged helium if excitation is included in the calculation [34].

We point out that the curve in figure 2 relies on the fact that the excited electron will be field ionized instantaneously. If this is the case then the curve depends on the field at the return and thus also on the laser power and the atom under consideration. The critical field for above the barrier ionization can be estimated using equation (4.1) in [35] to be 2.97×10^6 V m^{-1} for a binding energy of 11 eV (first excited state of Ar^{1+}). At much lower peak powers than used in

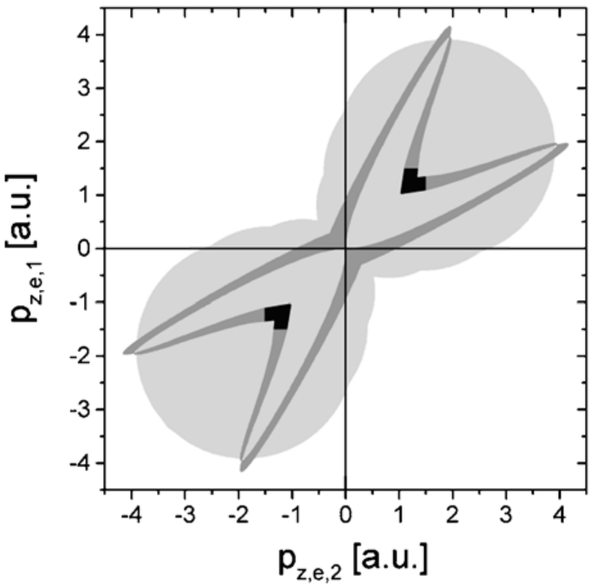

Figure 3. Classically allowed phase space within the rescattering model for 780 nm at intensity 4.7×10^{14} W cm^{-2}. The light grey bone-shaped area shows the ionization and the medium grey curve shows the locus of events for rescattering with excitation between the first excited state and the continuum, assuming instantaneous quenching. The black area indicates the region of the most likely rescattering phase.

this work one can estimate that there is a chance for at least the lowest excited states to survive a light cycle which will change the above scenario completely (see for higher fields [36]).

Electron impact excitation shows dominant resonant structure (see e.g. [37]). These resonances result from intermediate doubly excited states which autoionize. This is similar to the intermediate formation of an excited complex suggested by Sacha *et al* [26]. Once the returning electron has sufficient energy to ionize, the two electrons can share their energy arbitrarily in the collision and then both acquire additional momentum from the field. The field determines a maximum return energy (3.17 times the pondoremotive potential) and hence a maximum momentum for the electrons. This region of phase space classically allowed for ionization is shown by the shaded area. Most of the intensity in our measured distributions is well within this classically allowed phase space. Quantum mechanically even other regions of phase space become accessible since the intermediate state can be off the energy shell (see [27]). How the classically allowed phase space is populated depends on the details of the (e, 2e) process which is dominated by the long-range electron–electron repulsion. This determines the energy sharing and the angular correlation between the electrons.

The effects of electron repulsion on the pattern shown in figure 1 are more quantitatively explored in a recent model calculation using the S-matrix theory [27,38]. In these calculations the two different electron–electron potentials have been used for the recollision step: a (short-range) contact potential and a coulomb potential. For the contact potential, as it neglects the long-range final state repulsion, most of the electrons are found in one spot on a diagonal similar to that presented in figure 1(*d*). The full Coulomb potential, however, yields distributions with one fast and one slow electron dominating, similar to the experimental findings shown in figure 1(*a*). It remains puzzling that the inclusion of the correct Coulomb potential between the electrons yields worse agreement with the data integrated over all transverse momenta. Our argument presented through figure 3 suggests that this is due to the fact that these calculations do not include the excitation channel. Clearly a full calculation including all intermediate channels and the correct electron–electron repulsion is necessary to further elucidate this process. From our data we conclude that at larger transverse momenta, rescattering with excitation dominates, while at smaller transverse momenta the (e, 2e) ionization is dominant. In the latter channel electron–electron repulsion leads to the dominance of one fast electron accompanied by a slower one.

In conclusion we have seen the first evidence of the effect of electron repulsion in the final state for double ionization in strong fields. In addition we have identified contributions due to excitation and ionization in the rescattering process. Even though our present experiment has not provided a full picture of the three-body breakup (since for one of the electrons the transverse momentum remains unobserved) a complete mapping of the correlated final state momentum distribution should be within reach in the near future. In particular, experiments considering helium as a real three-body system and with its well-separated intermediate-ion excited states are promising candidates for a full exploration of the double ionization process in strong fields.

We are indebted to Horst Schmidt-Böcking for enthusiastic support of this project and Robert Moshammer and Joachim Ullrich for many helpful discussions. A Lahmam-Bennani and E Weigold gave valuable arguments for the interpretation of the recollision step. We thank W Becker, S P Goreslavskii and S V Popruzhenko for communicating their theoretical results prior to publication and A Becker for providing theoretical results for the calibration of our laser intensity. This work is supported by DFG, BMBF, GSI and DAAD. TW is grateful for the financial support of the Graduiertenförderung des Landes Hessen. RD acknowledges support from the Heisenberg-Programm of the DFG.

References

[1] Lambropoulos P, Maragakis P and Zhang J 1998 *Phys. Rep.* **305** 203
[2] Fittinghoff D N, Bolton P R, Chang B and Kulander K D 1992 *Phys. Rev. Lett.* **69** 2642
[3] Walker B, Sheehy B, DiMauro L F, Agostini P, Schafer K H and Kulander K C 1994 *Phys. Rev. Lett.* **73** 1227
[4] Larochelle S, Talebpour A and Chin S L 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 1201
[5] Guo C, Li M, Nibarger J P and Gibson G N 1998 *Phys. Rev.* A **58** R4271
[6] Becker A and Faisal F H M 1996 *J. Phys. B: At. Mol. Opt. Phys.* **29** L197
[7] Becker A and Faisal F H M 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** L335
[8] Becker A and Faisal F H M 1999 *Phys. Rev.* A **59** R1742
[9] Liu W C, Eberly J H, Haan S L and Grobe R 1999 *Phys. Rev. Lett.* **83** 520
[10] Watson J B, Sanpera A, Lappas D G, Knight P L and Burnett K 1997 *Phys. Rev. Lett.* **78** 1884
[11] Kuchiev M Yu 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** 5093
[12] Lappas D G and van Leeuwen R 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** L249
[13] LaGattuta K J and Cohen J S 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 5281
[14] Kulander K C, Cooper J and Schafer K J 1995 *Phys. Rev.* A **51** 561
[15] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[16] Weber Th, Weckenbrock M, Staudte A, Spielberger L, Jagutzki O, Mergel V, Urbasch G, Vollmer M, Giessen H and Dörner R 2000 *Phys. Rev. Lett.* **84** 443
[17] Moshammer R *et al* 2000 *Phys. Rev. Lett.* **84** 447
[18] Weber Th, Weckenbrock M, Staudte A, Spielberger L, Jagutzki O, Mergel V, Urbasch G, Vollmer M, Giessen H and Dörner R 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** L127
[19] Witzel B, Papadogiannis N A and Charalambidis D 2000 *Phys. Rev. Lett.* **85** 2268
[20] Lafon R, Chaloupka J L, Sheehy B, Paul P M, Agostini P, Kulander K C and DiMauro L F 2001 *Phys. Rev. Lett.* **86** 2762
[21] Moshammer R, Feuerstein B and Ullrich J 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** L823
[22] Becker A and Faisal F H M 2000 *Phys. Rev. Lett.* **84** 3546

[23] Kopold R, Becker W, Rottke H and Sandner W 2000 *Phys. Rev. Lett.* **85** 3781
[24] Lein M, Gross E K U and Engel V 2000 *Phys. Rev. Lett.* **85** 4707
[25] Bhattacharyya S and Mitra S 2000 *Phys. Rev.* A **62** 032709
[26] Sacha K and Eckhardt B 2001 *Phys. Rev.* A **63** 043414
[27] Goreslavskii S P and Popruzhenko S V 2001 *Opt. Express* **8** 395
[28] Weber Th, Giessen H, Weckenbrock M, Staudte A, Spielberger L, Jagutzki O, Mergel V, Urbasch G, Vollmer M and Dörner R 2000 *Nature* **404** 608
[29] Briggs J and Schmidt V 2000 *J. Physique* **33** R1
[30] Weber Th *et al* 2001 *Opt. Express* **7** 368
[31] Dörner R, Mergel V, Jagutzki O, Spielberger L, Ullrich J, Moshammer R and Schmidt-Böcking H 2000 *Phys. Rep.* **330** 96–192
[32] see www.roentdek.com
[33] Moshammer R, Unverzagt M, Schmitt W, Ullrich J and Schmidt-Böcking H 1996 *Nucl. Instrum. Methods* B **108** 425
[34] Bhardwaj V R, Aseyev S A, Mehendale M, Yudin G L, Villeneuve D M, Rayner D M, Ivanov M Yu and Corkum P B 2001 *Phys. Rev. Lett.* **86** 3522
[35] Protopatas M, Keitel C H and Knight P L 1997 *Phys. Rep.* **60** 389
[36] Feuerstein B *et al* 2001 *Phys. Rev. Lett.* at press
[37] Filipovic D, Marinkovic B, Pejcev V and Vuskovic L 1988 *Phys. Rev.* A **37** 356
[38] Goreslavskii S P, Popruzhenko S V, Kopold R and Becker W 2001 submitted