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FAST TRACK COMMUNICATION

Two-photon double ionization of Ne by free-electron laser radiation: a kinematically complete experiment

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Abstract
We present kinematically complete data on two-photon double ionization of Ne induced by short (∼25 fs) intense (∼5 × 10^{13} W cm^{-2}) free-electron laser pulses at 44 eV. The observed electron energy spectrum points to the dominance of 'sequential' ionization. We analyse state-selective angular distributions as well as the two-electron angular correlation function, and suggest a method to determine the time delay between both ionization steps. The measured angular asymmetry (β-) parameters significantly deviate from the results of an earlier non-coincident experiment providing benchmark data for theory.

(Some figures in this article are in colour only in the electronic version)

The emission of several electrons from atoms, molecules, clusters or solids as a consequence of the absorption of a single photon can only occur if the electrons interact with each other, i.e. due to an electron–electron correlation. One of its clearest, thus fundamental, realizations is *one-photon double ionization* (OPDI), explored over decades at synchrotrons [1] and culminating in recent fascinating fully differential results for a He atom [2] and H_2 molecule [3]. Tremendous progress in theory and the development of 'reaction microscopes' [4] allowing for the kinematically complete characterization of the above reactions have led to the commonly accepted view that OPDI is fully understood, at least for the most basic systems, He and H_2.

Two-photon double ionization (TPDI) instead became only accessible recently when intense EUV/XUV light sources, the free-electron lasers (FEL) [5] and high harmonics (HH) from infrared lasers [6] enabled first experiments, sparking enormous theoretical activities with controversial results for even the simplest reaction, TPDI of He (see e.g. [7–12]). Two channels are usually distinguished in TPDI. The sequential double ionization (SDI) (see the inset of figure 1(a)) is allowed if the photon energy $\hbar\omega$ is larger than the ionization potential I_P^+ of the singly charged ion. It is usually seen as

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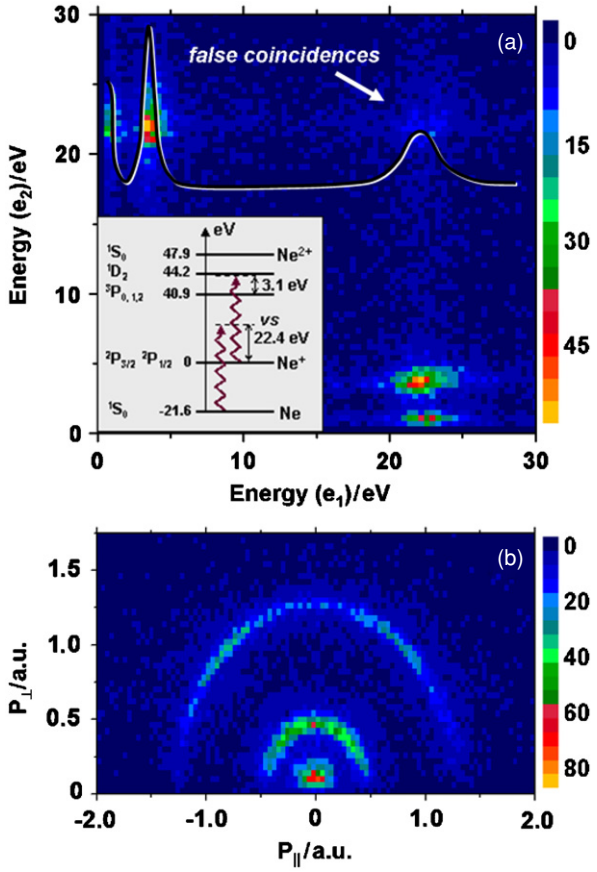

Figure 1. (a) Coincident two-electron energy spectrum for TPDI of Ne by 44 eV FLASH photons. The solid line shows singly differential electron energy spectrum. Inset: schematics of the relevant Ne energy levels and possible ionization pathways. (b) 2D electron momentum distribution for the same reaction. $P_{\parallel}$ denotes the momentum component parallel and $P_{\perp}$ perpendicular to the FLASH polarization direction. Note that in this representation, different emission angles are weighted with different solid angles.

proceeding sequentially in time with two independent photo absorption events: one from the atom and the other from the relaxed singly charged ion. The direct or 'non-sequential' double ionization (NSDI) instead, the only possibility at lower photon energies ($I_P^+ > \hbar\omega$ but $2\hbar\omega > I_P^+ + I_P$ with I_P being the ionization potential of the neutral), requires the 'instantaneous' absorption of the two photons through an intermediate virtual state (versus in the inset of figure 1(a)) and is considered to be highly correlated with an essentially smooth energy partition between the two electrons.

Whereas NSDI has received most attention until now, considerable theoretical work was recently dedicated to the sequential pathway [8, 9, 12–16]. Initially [13] SDI was characterized by several typical features. First, the electrons are considered to be emitted independent of each other. Second, whenever sequential TPDI is energetically allowed, it is expected to dominate the total cross-section. Third, the predicted photoelectron spectrum is characterized by two lines defined by energy conservation and the two ionization potentials. While the latter two expectations are in line with the results of recent more elaborate calculations [12, 14], the former one has been questioned, and an angular correlation between the emitted electrons has been predicted [16] pointing to the subtle nature of the process.

Until now, essentially none of these predictions for SDI have been confirmed experimentally. Ion-yield measurements, mostly available up to now [6, 17, 18], cannot even discriminate between NSDI and SDI since both exhibit the same quadratic intensity dependence [18]. Recent Ne^{2+} and He^{2+} momentum distributions measured at the *f*ree-electron *las*er at *H*amburg (FLASH) [19, 20] revealed distinctly different patterns for direct and sequential TPDI, clearly identifying them for the first time in qualitative agreement with theoretical predictions [9, 20] but providing no deeper insight. Similarly, although the first non-coincident photoelectron spectra for SDI of Ar and Ne manifested the lines expected for the second step in sequential TPDI [21], they could not exclude the presence of a contribution from the direct channel since the line due to the SDI appears on top of ∼10% background. Furthermore, they neither yielded information on the angular correlation between both electrons nor revealed angular distributions and β-parameters for the 'first' ionization step because the corresponding electrons appear at the same energy as those from single ionization (SI).

In this communication, we report on the first kinematically complete experiment on TPDI of Ne at the photon energy of 44 eV, delivered by FLASH. Measuring three-dimensional (3D) momentum vectors of the Ne^{2+} ion and of both emitted electrons in a triple coincidence, we observe a characteristically structured photoelectron spectrum proving that the ionization predominantly proceeds via stationary intermediate states of the Ne^+ ion, i.e. via the 'sequential' channel. Since our energy resolution was sufficient to distinguish events corresponding to different final states of the Ne^{2+} ion, we extracted state-selected angular asymmetry parameters (β-) from the measured angular distributions for both electrons. The β-parameters obtained lie well below one and in-between two other theoretical predictions, and deviate by a factor of three from the previous non-coincident experimental result at a slightly higher photon energy. Finally, exploring the angular correlation function of both electrons and comparing it with theoretical predictions, we discuss the role of the electron–electron correlation in a generally accepted picture of a stepwise SDI and suggest a method for a direct measurement of the time delay between the two ionization steps.

The present experiment has been performed using a setup similar to the one described in [19, 20]. Briefly, a 'reaction microscope' [4] was installed in the focus of the BL2 beam line of the FLASH. Linearly polarized radiation (28.2 ± 0.3 nm, 44 ± 0.5 eV) with the effective repetition rate of 60 Hz (structured in pulse trains with a repetition rate of 5 Hz, 12 pulses per train, pulse separation 10 μs, single pulse duration ∼25 fs) was focused onto a 30 μm spot within the collimated supersonic gas jet in the centre of the ultra-high vacuum ($<8\times10^{-12}$ mbar) chamber. Created ions and electrons were guided to position-sensitive channel plate detectors by weak electric (0.86 V cm^{-1}) and magnetic (10.8 G) fields. From the measured times of flight and positions on the detectors, the full momentum vectors of

emitted ions and electrons were reconstructed. A 4π acceptance solid angle has been achieved for ions and for electrons with kinetic energies up to 27.5 eV (see [4] for details). Electrons with kinetic energies larger than 27.5 eV initially flying towards the ion detector were not detected. The FEL polarization was parallel to the direction of the gas jet propagation, and the averaged value of the peak intensity estimated from the beam parameters corresponded to $\sim 5 \times 10^{13}$ W cm^{-2}. It should be noted that the present triple-coincidence experiment is at the absolute feasibility limit at the current repetition rate of FLASH, and the statistical significance of the data presented here was limited by the available beam time under extraordinarily competitive boundary conditions.

Figure 1(a) depicts the measured coincident two-electron energy distribution integrated over all emission angles (with the solid line showing the total singly differential energy spectrum). Here, only those events for which the sum-momentum of all emitted fragments is zero for each Cartesian coordinate are included. Apart from a small contribution from false coincidences (indicated by the arrow), the correlated energy spectrum consists of four distinct peaks symmetric with respect to the main diagonal. For each pair of peaks, the energy of the electron from the first ionization step is centred at 22.5 eV, which is identical to the measured energy of electrons from SI. The second electron either emerges with a kinetic energy of about 3 eV, or with nearly zero energy, indicating that the Ne^{2+} ion is left in the $^3P_{0,1,2}$ state (40.9 eV with respect to the Ne^+ ground state, triplet fine structure is not resolved), or in the $^1D_2$ state (44.2 eV), respectively. The $^1S_0$ final state (47.9 eV) is energetically not accessible for TPDI. The contributions from direct TPDI, which would be distributed along the lines parallel to the second diagonal [11], as well as the double ionization events induced by second or third harmonics are not observed in the spectrum.

Figure 1(b) displays the 2D momentum distribution of the electrons originating from double ionization. One can clearly observe three structures corresponding to the different electron energies in figure 1(a). The outer half-ring corresponds to the 22.5 eV electrons from the first ionization step, the middle one with the radius of ~0.5 au reflects those with ~3eV energy from the second step leading to the ^3P final state of Ne^{2+} and the spot close to zero originates from the second step low-energy electrons corresponding to the ^1D final state.

From the measured three momentum components for each of the electrons, we reconstruct the angular distributions of the emitted electrons, and extract angular asymmetry (β) parameters. The angular distribution for single ionization has been fitted with the standard expression for a dipole-type emission:

$$\frac{d\sigma(E)}{d\Omega} = \frac{\sigma(E)}{4\pi}\left(1 + \beta_2^{1+}(E) P_2(\cos\theta)\right), \qquad (1)$$

where E is the photon energy, $\sigma(E)$ is the partial photoionization cross-section, θ is the electron emission angle with respect to the FEL polarization direction and P_2 is the second Legendre polynomial. We obtain $\beta_2^{1+} = 0.92 \pm 0.01$, in good agreement with the literature values for this photon energy [22]. In the case of double ionization, for which the angular distribution is given by the higher order Legendre polynomial since this is a two-photon process [16]:

$$\frac{d\sigma(E)}{d\Omega} = \frac{\sigma(E)}{4\pi} \times \left(1 + {}^{(i)}\beta_2^{2+}(E) P_2(\cos\theta_i) + {}^{(i)}\beta_4^{2+}(E) P_4(\cos\theta_i)\right), \qquad (2)$$

we obtain the values of ${}^{(1)}\beta_2^{2+} = 0.84 \pm 0.08$ and ${}^{(1)}\beta_4^{2+} = -0.12 \pm 0.11$ for the first ionization step ($i = 1$), and ${}^{(2)}\beta_2^{2+} = 0.46 \pm 0.08$ and ${}^{(2)}\beta_4^{2+} = -0.14 \pm 0.1$ for the second one ($i = 2$). Only the events where the Ne^{2+} ion is left in the ^3P final state are considered in both cases.

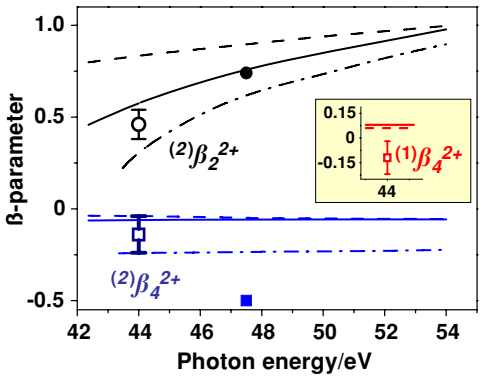

Figure 2. Measured and calculated angular asymmetry parameters ${}^{(2)}\beta_2^{2+}$ (black) and ${}^{(2)}\beta_4^{2+}$(blue) as a function of photon energy. Inset (red): ${}^{(1)}\beta_4^{2+}$-parameter for the electrons from the first step of TPDI. Open symbols: present experiment; full symbols: experiment [21] (taken from [15]); solid lines: MCHF calculation (this work); dashed lines: MCDF calculation (this work); dashed-dotted lines: HF calculation of Kheifets [15].

Figure 2 shows the comparison of the obtained ß-values (open symbols) with the theoretical predictions. Solid and dashed lines represent the results of the present multi-configurational Hartree-Fock (MCHF) and Dirac-Fock (MCDF) calculations, respectively. The details of both calculations can be found in [16]. For a correct direct comparison, the theoretical results were averaged over the unresolved $^3P_{0,1,2}$ final states of the Ne^{2+} ion (see [16]). The predicted state-selective ${}^{(2)}\beta_2^{2+}$ parameters considerably deviate from each other. Our experimental value lies much closer to the MCHF result, which is slightly above the upper limit of the experimental error. Notably, recent calculations by Kheifets [15] (shown as a dashed-dotted line in figure 2) predict an even lower value, thus, placing our experimental point exactly in-between these two theoretical curves. A similar behaviour can be observed for the ${}^{(2)}\beta_4^{2+}$ parameter. Here, both MCHF and MCDF calculations yield almost identical values, again lying at the upper limit of the experimental error bar, whereas the results of [15] fit the lower one. Thus, while we cannot discriminate between different theories for ${}^{(2)}\beta_4^{2+}$, we found that the MCDF calculation clearly overestimates ${}^{(2)}\beta_2^{2+}$.

In [15], the theoretical predictions were compared with the values of ${}^{(2)}\beta_2^{2+}$ and ${}^{(2)}\beta_4^{2+}$ parameters obtained in the non-coincident experiment of Braune *et al* at 47.5 eV [21], shown as full symbols without error bars in figure 2. Whereas the ${}^{(2)}\beta_2^{2+}$ value agrees well with the prediction of the MCHF calculation,

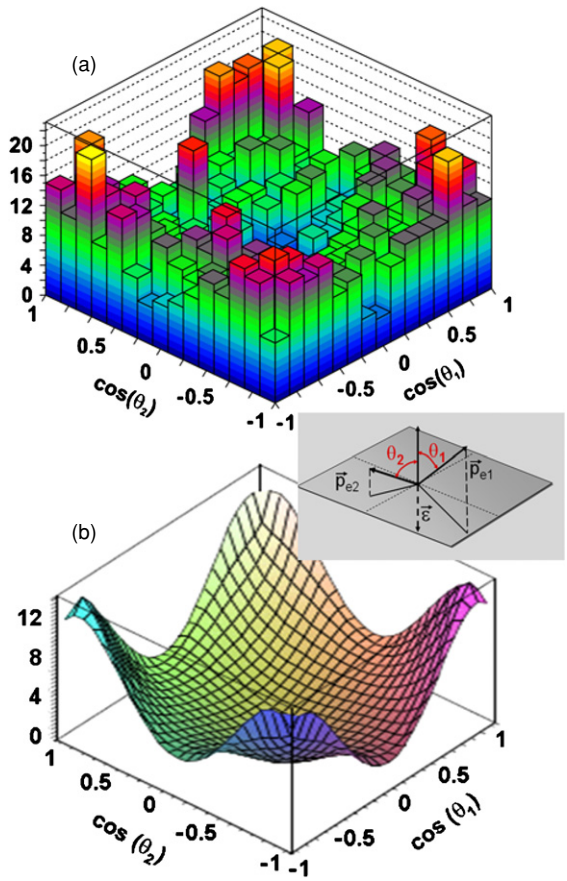

Figure 3. Experimental (a) and theoretical (b) probability density distribution of two emitted electrons as a function of ($\cos(\theta_1)$, $\cos(\theta_2)$), where θ is the emission angle with respect to the FLASH polarization direction. Inset: sketch of the experimental geometry.

and, accounting for the expected wavelength dependence, also with our experimental result, the one for ${}^{(2)}\beta_4^{2+}$ is much lower than all three theoretical values. Correspondingly, it also considerably differs from the value obtained in the present experiment. Since none of the three calculations predicts significant wavelength dependence for ${}^{(2)}\beta_4^{2+}$ in this energy range, the reasons for this discrepancy remain unclear.

Up to now, we have considered the electron angular distributions for the second ionization step independently. However, as discussed in [16], one might expect an angular correlation between the electrons, mainly as a consequence of the polarization of the intermediate ionic state. In order to explore this, we plot in figure 3(a) the measured emission angle of the first electron θ_1 versus that of the second electron θ_2, and compare it with the theoretical (MCDF) angular correlation function (figure 3(b)). We observe an overall similar pattern for both theory and experiment. Nevertheless, the slight but significant asymmetry of the theoretical plot revealing the angular correlation, i.e. a change of the angular distribution of one electron as a function of the emission direction of the second, cannot be clearly identified in the experimental results within the present statistical significance.

However, since according to the theoretical expectations [16] both electrons in sequential TPDI are not emitted independently, this might also be imprinted in the angular distribution of the first step electrons, which then should differ from the one obtained for SI. We do observe some difference reflected in the non-zero value of ${}^{(1)}\beta_4^{2+}$, slightly outside the statistical error bar (${}^{(1)}\beta_4^{2+} = -0.12 \pm 0.11$, red data point in the inset of figure 2(b)). Analysing this effect in the framework of the MCHF and MCDF approaches, we obtain non-zero values of ${}^{(1)}\beta_4^{2+} = 0.08$ and ${}^{(1)}\beta_4^{2+} = 0.06$, respectively (red full and dashed lines in the inset of figure 2). Thus, even though the observed deviations from zero are weak, we do find a discrepancy between theory and experiment beyond the statistical experimental error. Since the deviations are rather small and in view of the fact that both theoretical approaches differ substantially in the prediction of ${}^{(2)}\beta_2^{2+}$, speculations about possible reasons for that might not seem adequate. Nonetheless, as discussed in [16] depolarization of the intermediate Ne^{1+} consisting of a coherently excited superposition of the ${}^2P_{3/2}$ and ${}^2P_{1/2}$ states might be one physical reason neglected in the present theories.

In summary, we have presented the first fully differential data for a two-electron nonlinear process in the XUV domain, TPDI of Ne. We undoubtedly observe the spectrum predicted for the 'sequential' mechanism, without visible contributions from the competing direct channel. From the 3D momentum distributions of the two electrons measured in coincidence, we extract the angular asymmetry parameters for both steps, which, being in good overall agreement with two of the available theoretical predictions and discarding one of them, allow one to benchmark theory for this very fundamental reaction. For the ${}^{(2)}\beta_4^{2+}$ parameter, both our experimental and theoretical results show an unexpectedly large discrepancy with the non-coincident measurement of Braune *et al* [21] at somewhat higher photon energy, and it remains to be clarified whether this deviation is due to an experimental error, or originates from some unexpected wavelength dependence overlooked by theory.

Finally, it should be noted that possible modification of the angular distribution for the first step electrons compared to single ionization raises a very general question concerning the nature of sequential TPDI, since it appears rather counterintuitive that the second electron, which might be set free much later within the FEL pulse, influences the emission characteristics of the first one. This brings one back to the conclusion stated in the initial paper on the subject [13], claiming that 'there is no way of distinguishing sequential from simultaneous', i.e. the timing of the electron emission cannot be revealed from the experimental data. However, latest advances of the FEL technology suggest a way to directly measure a potential time delay between the two emission steps via a two-colour 'streaking' pump probe experiment. Similar to the concept of the 'attosecond streak camera', the energy of the electrons from TPDI can be modulated by an infrared field yielding the time when each electron was set free [23]. The streaking field needs to be fully synchronized with the ionizing XUV pulse, and its half-cycle has to be longer than the XUV pulse duration. Such a scheme can now be realized exploiting a novel THz beamline at FLASH, where streaking at wavelengths up to 93 μm has recently been demonstrated [24]. An intriguing question is whether such a measurement would change the angular correlation function of both electrons.

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References

[1] Avaldi L and Huetz A 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** S861
[2] Knapp A *et al* 2002 *Phys. Rev. Lett.* **89** 033004
[3] Akoury D *et al* 2007 *Science* **318** 949
[4] Ullrich J *et al* 2003 *Rep. Prog. Phys.* **66** 1463
[5] Ackermann W *et al* 2007 *Nat. Photonics* **1** 336
[6] Hasegawa H *et al* 2005 *Phys. Rev.* A **71** 023407
[7] Nikolopoulos L A A and Lambropoulos P 2007 *J. Phys. B: At. Mol. Opt. Phys.* **40** 1347
[8] Horner D A *et al* 2007 *Phys. Rev.* A **76** 030701(R)
[9] Horner D A, Rescigno T N and McCurdy C W 2008 *Phys. Rev.* A **77** 030703(R)
[10] Foumouo E *et al* 2008 *J. Phys. B: At. Mol. Opt. Phys.* **41** 051001
[11] Feist J *et al* 2008 *Phys. Rev.* A **77** 043420
[12] Guan X, Bartschat K and Schneider B I 2008 *Phys. Rev.* A **77** 043421
[13] Bachau H and Lambropoulos P 1991 *Phys. Rev.* A **44** R9
[14] Laulan S and Bachau H 2003 *Phys. Rev.* A **68** 013409
[15] Kheifets A 2007 *J. Phys. B: At. Mol. Opt. Phys.* **40** F313
[16] Fritzsche S, Grum-Grzhimailo A N, Gryzlova E V and Kabachnik N M 2008 *J. Phys. B: At. Mol. Opt. Phys.* **41** 165601
[17] Benis E P *et al* 2006 *Phys. Rev.* A **74** 051402(R)
[18] Sorokin A *et al* 2007 *Phys. Rev.* A **75** 051402(R)
[19] Moshammer R *et al* 2007 *Phys. Rev. Lett.* **98** 203001
[20] Rudenko A *et al* 2008 *Phys. Rev. Lett.* **101** 073003
[21] Braune M, Reinköster A, Viefhaus J, Lohmann B and Becker U 2007 HASYLAB Annual Report, p 457; http://hasyweb.desy.de/science/annual_reports/2007_report/part1/contrib/40/21686
[22] Schmidt V 1986 *Z. Phys.* D **2** 275
[23] Itatani J *et al* 2002 *Phys. Rev. Lett.* **88** 173903
[24] Gensch M *et al* 2008 *Infrared Phys. Technol.* **51** 423
Fruehling U *et al* 2009 to be published