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**Abstract.** In recent combined experimental and theoretical study we have explored non-sequential double ionization of neon and argon atoms in the infrared light field (800nm) below the recollision threshold. We find that the two-electron correlation dynamics depends on atomic structure– "side-by-side emission" (correlation) for Ne and "back-to-back emission" (anticorrelation) for argon atoms. This can be explained theoretically within our three dimensional classical model calculation including tunnelling effect. The multiple recollisions as well as recollision-induced-excitation-tunnelling (RIET) effect dominate the anticorrelation of argon, whereas the laser-assisted instantaneous recollision dominates the correlation of neon.

## 1. Introduction

Within the past three decades extensive works on the interaction of intense laser fields with atoms and molecules have resulted in a profound understanding of various strong-field phenomena, e.g., above-threshold ionization [1], high-harmonic generation [2] and nonsequential double ionization (NSDI) [3,4]. Few-electron correlation is most important in NSDI. Until the present day, it still resists any comprehensive modelling. NSDI not only manifested its importance in the enhancement of double charged ions at moderate laser intensities [4] and but, represents an outstanding subtleties of electron-electron correlations in strong fields. Strong-field ionization mechanisms of single ionization are often classified by the Keldysh [5] parameter $=(I_p/2U_p)^{1/2}$ ($I_p$: ionization potential; $U_p = E_0^2/\omega^2$:pondermotive potential with $E_0$ the laser field amplitude and $\omega$ its frequency; atomic units (a.u.) are used throughout the paper). For $< 1$, at moderately large intensities, tunnelling ionization is dominant. Eventually, for $\ll 1$ and very strong fields the potential might be such strongly suppressed that the electrons can just escape "over the barrier". Vice versa, multi-photon processes are expected to dominate at smaller intensities and $> 1$. NSDI essentially occurs in the tunnelling ionization regime, below the over barrier ionization regime and is usually explained within the quasiclassical re-collision model [6-8].

Here, the first electron is set free near the maximum of laser field, accumulates energy (up to 3.17 $U_p$) during about half a laser cycle, is thrown back on its parent ion near a zero-crossing of the laser field and, in a inelastical collision, kicks out the second electron. As shown in many experiments and calculations, both emitted electrons prefer to move into the same hemisphere along the laser polarization axis [9-14]. Even though this general scenario is well established, distinct differences do occur for different target species, i.e., significant structure dependence is observed for NSDI. Up to now, this is qualitatively explained via target dependent cross sections for (re-colliding) electron-impact excitation of the ionic core: If this cross section is small, direct re-collision ionization dominates with both electrons being mainly ejected into the same hemisphere. Is it large, as, e.g. for Ar 3s to 3p excitation, re-collision-induced excitation with subsequent field ionization [10] becomes important, with both electrons being equally likely emitted in the same or opposite hemisphere. One should note, however, that this discussion cannot be considered completely settled, since other ionization mechanisms leading to anti-correlation at high intensities, close to the over barrier ionization regime, have been discussed recently [15].

One intriguing though still widely unexplored question did arise early on within above recollision picture. What does happen when the energy of the recolliding electron of up to 3.17 $U_p$ is not sufficient to directly ionize the parent ion or, at lower energies, not even to reach its first excited state? This is a situation, where previous classical considerations do not hold any more and where one enters the fundamentally important non-perturbative multi-photon – multi-electron regime. Still, multiple ionization rates were found to be dramatically enhanced due to electron-electron correlation. At low intensities below the re-collision threshold, where one enters the multi-photon ionization regime, anti-correlation between the electrons from NSDI of argon has been observed recently [16] and new ionization mechanisms have been suggested [17]. In this contribution, we investigate electron correlation in NSDI and its target-species dependence for Ne and Ar at lowest intensities ever addressed, below the recollision threshold, even closing to the double ionization threshold. In kinematically complete measurements we find opposite correlation behaviour in the two-electron momentum distributions along the polarization direction, dominating emission into opposite hemispheres (back-to-back emission) for Ar (so-called anti-correlation) and into the same hemisphere (side-by-side emission) for Ne (so-called correlation), which we partly explain within three-dimensional classical calculations including tunnelling of the second electron. Inspecting the transverse momentum distributions we shed light on the dynamics, investigating Coulomb focusing effects, and find non-classical dynamics for the NSDI of Ne in threshold regime.

Dedicated classical model calculation [18] on the other hand, considering the contributions from multiple re-collisions and, in addition, implementing tunnelling of the second electron [17], clearly revealed the back-to-back emission characteristics observed in the experiment. Whereas in [21], the anti-correlated electrons were found to be mainly produced via multiple collisions in the first or second field maximum after re-collision, the analysis in [17] yields a more subtle picture. Here, the contributions to anti-correlated electron emission due to classical multiple recollisions induced direct-ionization can be distinguished from those due to recollision induced excitation plus tunneling (RIET) of the second electron. Whereas the recollision induced direct ionization contribution to correlated emission decreases with decreasing intensity, the RIET part is expected to increases. For anti-correlated events, instead, both mechanisms contribute when lowering the intensity making anticorrelation to dominate over correlation deep in the multiphoton ionization regime in agreement with experiment. Currently, quantum calculations are widely lacking below the recollision threshold and the dependence on the target structure, expected to be very important for the case of excitation-tunnelling ionization, has not been investigated at all.

## 2. Experimental setup

The coincident detection of electrons and ions of NSDI in the multiphoton ionization regime is challenging due to the exceedingly low double ionization yields. The major breakthrough enabling such measurements was via the demonstration of a high-power (peak power >16MW) femtosecond laser system approaching microjoule with unprecedented repetition rate (~6 MHz) [19,20]. The laser pulse duration is about 46fs. The laser pulse was tightly focused to about 1μm diameter and a
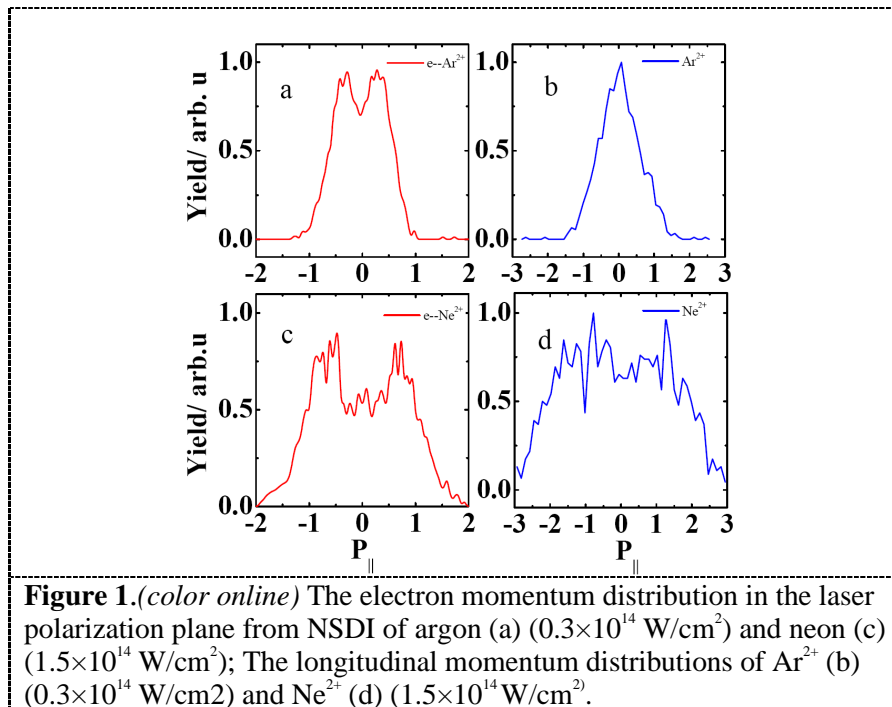
maximum intensity more than $2 \cdot 10^{14}$ W/cm$^2$ on the target could be achieved. Charged fragments, i.e., ions and electrons, were guided onto two position-sensitive channel plate detectors by weak electric (2V/cm) and magnetic (4.5 G) fields applied along the laser polarization axis. The full momentum vectors of the coincident recoil ions and electrons were calculated from their times of flight (TOF) and hitting positions on the detectors. The gas-jet density of reaction system is about $1 \cdot 10^{12}$/cm$^3$. The coincidence method, the especially designed reaction microscope, the momentum resolutions achieved for the various fragments as well as the intensity calibration procedure and accuracy are described in detail elsewhere [21, 22]. Under present settings, data collection for each target took several weeks in order to achieve reasonable statistical significance, very much at the limit of what is technically feasible. In the experiments, we recorded a total of 1780 double coincidence events (one ion and one electron) for Ne$^{2+}$ with a ratio of Ne$^{2+}$/Ne$^+$ ~ $1.5 \cdot 10^{-4}$ at an intensity of $1.5 \times 10^{14}$ W/cm$^2$ and 1160 counts for Ar at $3 \cdot 10^{13}$ W/cm$^2$ with Ar$^{2+}$/Ar$^+$ ~ $8 \cdot 10^{-4}$.

## 3. Experimental results and model calculations

The electron longitudinal momentum distributions from double ionization of argon and neon are illustrated in Figs. 2(a) and (c). Both of them show double-hump structure, but they are resulted from different mechanism. Figures 2b and 2d show the momentum distributions of double charged argon and neon. At higher intensities, the longitudinal momentum spectra of double charged ions show so pronounced double-hump structure [12,13]. The longitudinal momentum distributions of Ar$^{2+}$ and Ne$^{2+}$ show very different feature in the NSDI threshold regime. It is found that the longitudinal momentum distribution of Ar$^{2+}$ shows a sharp cusp-like structure. This is because large amount of the coincident electrons moving in opposite direction. The pronounced "double-hump" structure of the longitudinal momentum distribution at higher laser intensity completely disappears. Thus, the momentum distribution of double charged argon can not be predicted from simple re-collision picture in the multi-photon ionization regime. In contrast to argon, the momentum distribution of double charged neon is much broad because the two ionized electrons still prefer to move into the same hemisphere. However the double hump structure is not pronounced for the longitudinal momentum distribution of Ne$^{2+}$ in the multi-photon ionization regime.

The momentum cut-off is about $\pm(10Up)^{1/2}$ (~ 5Up for energy cut-off) for argon. This is consistent with the observation that the high-energy part of single ionization and the sum of the energies of both electrons from double ionization of argon show similar shape [9]. As to double ionization of neon, there is very little excess energy during the recollsion process in the threshold regime. The electron momentum from double ionization of neon gives a $\pm(2Up)^{1/2}$-cut-off (~2Up for energy cut-off) and is consistent with "momentum constraint" [10].

In order to understand the influence of atomic structure on NSDI below recollision threshold, we have performed three-dimensional semi-classical model calculations, in which the recollision-induced excitation-tunneling (RIET) effect has been taken into account within the Wentzel-Kramers-Brillouin (WKB) approach [17]. Considering the cross sections for (re-colliding) electron-impact excitation of the ionic core, the recollision-induced excitation effect should be reduced more for Ne than that of Ar. Thus, RIET effect is less important in the case of Ne. The energy level of the first excitation state of Ne$^+$ and the field-free double ionization threshold are much higher than those of argon. It is thus much more difficult to resonantly excite the second electron during re-collision in a more reasonable quantum-mechanical description. To take this effect approximately into account in our classical model, we have abandoned all double ionization trajectories for Ne if the inner electron is excited to an energy that is lower than the first excited state.

**Figure 1**.*(color online)* The electron momentum distribution in the laser polarization plane from NSDI of argon (a) $(0.3\times10^{14}$ W/cm$^2)$ and neon (c) $(1.5\times10^{14}$ W/cm$^2)$; The longitudinal momentum distributions of Ar$^{2+}$ (b) $(0.3\times10^{14}$ W/cm2) and Ne$^{2+}$ (d) $(1.5\times10^{14}$W/cm$^{2)}$.

The calculated longitudinal momentum distributions of electrons and double charged ions are shown in Fig. 2. All of concerned results agree with the experimental data very well. The classical ensemble model can present the explanation of the production of significant numbers of double ionization electrons with energy much above $2Up$ for argon. In almost all trajectories leading to high energy, at least one electron remains bound for at least a portion of a laser cycle after recollision. During the interaction the electron which will achieve high energy experiences a sign change of momentum in the laser polarization plane. The anticipated process of nuclear scattering of a free electron at recollision is involved. The another process is that the bounded electron changes sign of momentum after the recollision, escapes over a suppressed barrier, and achieves high energy in the complex interaction. In this case, both electrons prefer to emit into opposite direction. In both processes, there is a change of phase of electron oscillation relative to the laser phase, thus electron achieve high energy beyond $2Up$.



**Figure 2**. *(color online)* Same as Figure 1 but for model calculations

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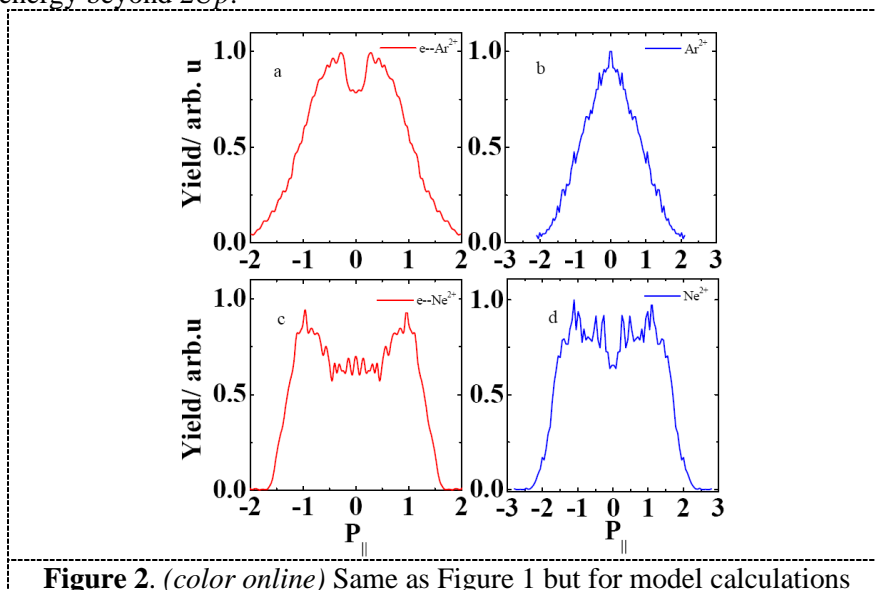
The correlation behaviours of Ar and Ne are also quite different in the threshold regime [23]. Even though the absolute single ionization count rate is rather comparable for both targets, the correlation maps as well as $P_\perp$-distributions show distinctly different shapes, as shown in Fig. 3. For argon a substantial amount of anti-correlated electrons are observed in line with our previous measurements at slightly higher intensity [5] and the correlated emission is found to dominate in case of neon!

Differences in the dynamics become especially obvious if one inspects the transverse momenta of electrons coincident to double ionization (red curve, right row) with a cusp-like structure for Ar that is substantially broadened in case of Ne. Even taking into account an at least 30 % uncertainty in the absolute determination of the intensity and, thus, thinking we might be much deeper in the multiphoton ionization regime for Ar than for Ne, we still find similarly distinct differences if we compare the Ne spectrum to the one for Ar at $7 \cdot 10^{13}$ W/cm$^2$ (dashed red line in Fig. 3b).

Inspecting the electron transverse momentum distributions for single ionization and double ionization for both target species, as shown in Figs 3(b) and 3(d) for the experiment as well as in Figs 4(b) and 4(d) for our model calculations we find in general pronounced "cusp-like" structures as observed before. It has been proposed that Coulomb focusing [24] or the Coulomb singularity [25] are responsible for their formation for single ionization in accordance with the present data (blue curves in Figs 3(b), 3(d), 4(b) and 4(d)). Whereas the experimental momentum distribution of electrons coincident with Ar$^{2+}$ ions (red line) also shows a clear cusp-like shape, only slightly broader than for single ionization and in good agreement with theory the respective distribution for Ne is significantly broader than predicted. In general, the cusp-like shape is obtained as a result of the singularity in the electron continuum wavefunction at zero energy in the presence of a Coulomb potential [21]. Thus, we conclude that for Ar double ionization both electrons have very little energy in the final state. This goes very much along with what would be expected within the RIET picture, where the second electron tunnels, being essentially set into the continuum with zero momentum and independently from the first electron, at another maximum in the laser field. Accordingly, our model calculations (Fig. 4 (b)) largely reproduce the experimental data in Fig. 3(b).
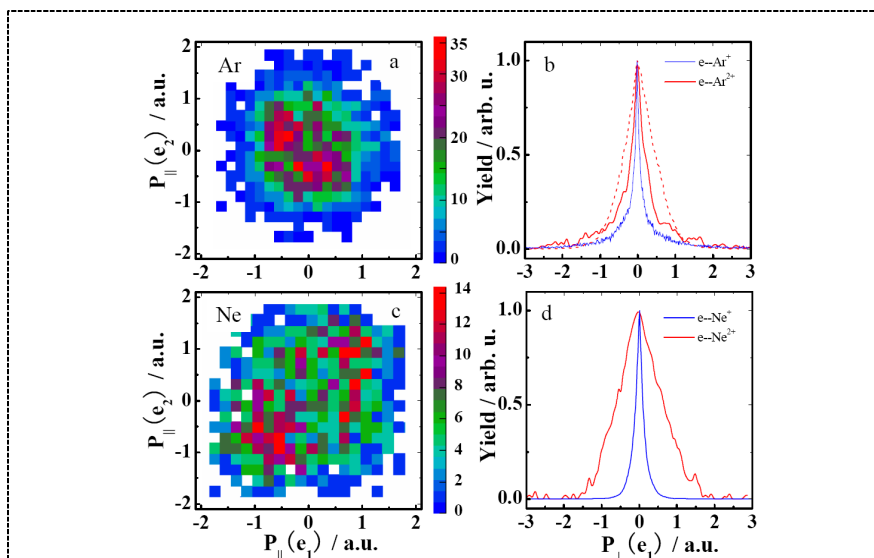


**Figure 3.** *(color online)* Parallel momentum correlation of two electrons for argon (a) ($3 \cdot 10^{13}$ W/cm$^2$) and neon (c) ($1.5 \cdot 10^{14}$ W/cm$^2$). b): Transverse momentum distributions of electrons in coincidence with Ar$^{1+}$ (blue line) and Ar$^{2+}$ at $3 \cdot 10^{13}$ W/cm$^2$ (solid red line) and at $7 \cdot 10^{13}$ W/cm$^2$ (dashed solid line) as well as d) for Ne$^{1+}$ and Ne$^{2+}$ at $1.5 \cdot 10^{14}$ W/cm$^2$.(experiment)

On the other hand electrons emerging coincident to $Ne^{2+}$ production are observed to receive much larger transverse momenta and show a more Gaussian-like shape. This indicates that the recollision mechanism still dominates, where the re-colliding electron could be scattered to a larger angle with it's longitudinal momentum being transferred to the transverse direction. Surprisingly, such a feature is not found in our classical model calculations where the transverse momentum distribution of an electron coincident to the simulated $Ne^{2+}$ ion, excluding trajectories that lead to unphysical low excitation energies, is shown in Fig. 4 (d). Obviously such a model does not capture the full quantum dynamics of the system in all of its aspects correctly, underlining the quest for quantum calculations.

In order to confirm the different correlation behaviour and to possibly even further penetrate into the multiphoton ionization regime, during the experiments, we have continued to decrease the laser intensity to about $1.5 \cdot 10^{13} \, W/cm^2$ for Ar and $8 \cdot 10^{13} \, W/cm^2$ for Ne. Here, however, we could not find any indication of double ionization events while running the measurements for several weeks for each species. This means that double ionization rates decrease by at least three orders of magnitude between $1.5 \cdot 10^{14} (3 \cdot 10^{13}) \, W/cm^2$ and $8 \cdot 10^{13} (1.5 \cdot 10^{13}) \, W/cm^2$ for neon (argon). The anticorrelation for neon was not able to be observed because of its extremely narrow transition region from correlation to anticorrelation, as discussed in [23].
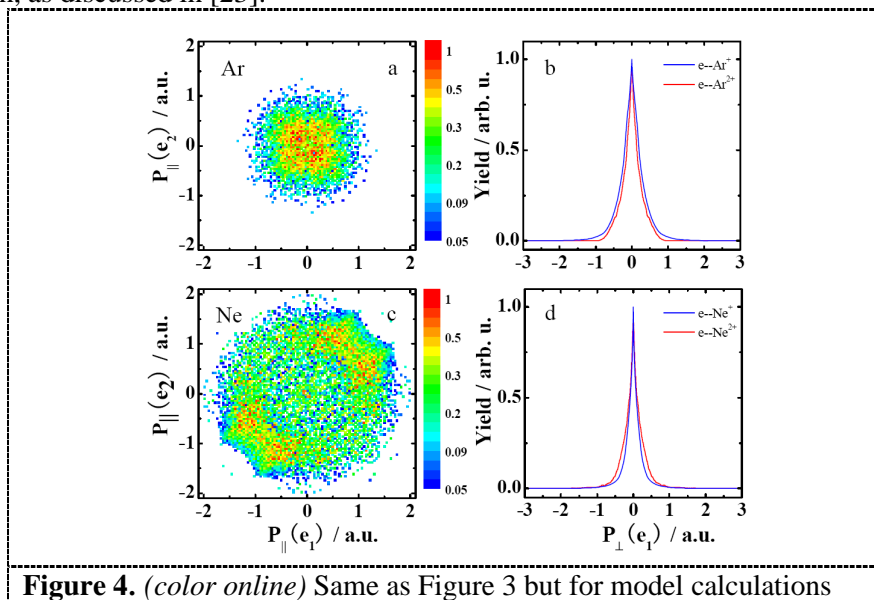


**Figure 4.** *(color online)* Same as Figure 3 but for model calculations

## 4. Conclusion

In this contribution, we present kinematically complete measurements and model calculations for NSDI of Ar and Ne in infrared laser fields below recollision threshold, at the lowest intensities ever investigated. Whereas for Ar anti-correlation (back-to-back emission) in the longitudinal two-electron momentum spectra is observed in agreement with previous measurements at slightly higher intensity, correlation (side-by-side emission) dominates for the Ne target similarly close to the threshold. Further lowering the intensity led to the complete absence of any double ionization event over weeks of data taking time. We can reproduce that salient feature in the correlation plot with 3D classical calculations including tunnelling for the second electron if, for the Ne target, all trajectories are rejected with re-collision energies below the first excitation level of $Ne^+$. This provides strong evidence that the target structure, i.e. the low-lying excited levels in $Ar^+$ along with the large electron-impact excitation cross section are the reason for this observation.

The electrons from of $Ne^{2+}$ can obtain larger transverse momentum and show more Gaussian shape. Under condition that the recollision mechanism dominates, the re-colliding electron could be in-elastically scattered to larger angle and obtain larger momentum. The transverse electron momentum of from $Ar^{2+}$ shows cusp-like structure. The effect of Coulomb potential of the parent ion is responsible to this observation. The Coulomb interaction between the electron and the ion reduces the

transverse spread of the electronic wave packet and shows strong focusing effect for argon. The Coulomb focusing effect leads to a small deflection of large impact parameter electrons whenever they approach the parent ion and enhance the probability for the electron to make a low impact parameter inelastic collision with the ionic core after multiple recollisions. This could be the intrinsic mechanism of sub-threshold re-collision process.

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