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Publisher's version / Version de l'éditeur:

<https://doi.org/10.1103/PhysRevLett.95.203003>

Physical Review Letters, 95, 20, 2005-11-10

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Controlling Attosecond Double Ionization Dynamics via Molecular Alignment

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(Received 5 April 2005; revised manuscript received 15 August 2005; published 10 November 2005)

We investigate the dynamics of double ionization in aligned nitrogen molecules. An ultrashort, weak laser pulse creates an aligned ensemble of molecules that is ionized with a subsequent, strong probe pulse. We find that the two electrons involved in nonsequential double ionization more likely exit the molecule in the same direction if it is parallel to the probe laser polarization, indicating that they are ejected within a few hundred attoseconds of each other. Double ionization is less probable and takes longer for perpendicular molecules.

DOI: [10.1103/PhysRevLett.95.203003](https://doi.org/10.1103/PhysRevLett.95.203003)

PACS numbers: 33.80.Rv, 34.80.Gs, 34.80.Pa

Experimental optical and collision physics developed relatively independently until the discovery that strong laser fields can force an ionizing electron to recollide with its parent ion [1]. Since then, collision physics has had a large impact on optics, solving major, long-standing problems. For example, high harmonic generation [2] of the driving laser field provides a tabletop source of coherent XUV radiation extending to more than 1 keV [3]. In addition, attosecond optical pulses, created by controlling the recollision electron [4], have extended the time resolution of ultrashort pulse technology by a factor of ~ 20 .

Optics is also having an impact on collision physics. Fast collisions are routinely used to generate subattosecond virtual photon fields that probe electron dynamics [5,6]. However, conventional collision experiments do not allow for pump-probe measurements to investigate attosecond dynamics, as the correlation of two collision events cannot be controlled. Recollision synchronizes electron collisional excitation with respect to the laser field, paving the way to exploit the attosecond duration of the recolliding electron burst [7]. The strong time-dependent electric field that drives the collision subsequently acts as a phase-locked streaking field to the electrons [8] and time resolves subsequent attosecond dynamics [9,10]. In addition, optical spectroscopic methods allow us to excite, orient, or align the collision partners before the collision occurs. This makes it possible to measure the influence of molecular geometry or excitation on attosecond multielectron dynamics.

We exploit both advances. We use an ultrashort, weak alignment pulse to create an ensemble of aligned, field-free nitrogen molecules [11] that we ionize with a delayed, intense “probe” pulse. By changing the time delay, we control the physical structure of the potential in which a recollision occurs. We detect the three-dimensional momenta of the two electrons from the following nonsequential double ionization [12–14] in coincidence and deduce the emission times of the electrons from the streaking properties of the phase-locked ionization field.

We demonstrate that attosecond double ionization dynamics is changed qualitatively by changing molecular alignment. Thus, the spatial structure of the scattering potential has a major effect not only on the probability, but also on the relative time of emission of the correlated electrons. The time difference is greater when the molecule is aligned perpendicular to the polarization of the ionizing laser than when it is aligned parallel.

Our results relate to experiments where attosecond double ionization dynamics has been retrieved from electron correlation [9]. They also relate to experiments on nonsequential double ionization of N_2 and O_2 molecules [13] where it has been shown that the electronic structure influences the electron momentum correlation.

For the experiment, we used ultrashort pulses from a Ti:sapphire regenerative amplifier (~ 40 fs, 800 nm, 30 kHz, 5 μ J). They were fed through a Mach-Zehnder interferometer to produce two pulses with adjustable delay. We use polarizers to split and recombine the beams, avoiding the 50% loss inherent to an interferometer with dielectric beam splitters. A half-wave plate in front of the interferometer determines the splitting ratio in its two arms. The time difference between the two pulses was controlled with a delay line.

Both pulses were focused into a supersonic gas jet with an $f = 50$ mm, $f/2$ parabolic mirror. For the gas jet, N_2 at a backing pressure of 2 bar was introduced through a 30 μ m nozzle into a differentially pumped source chamber. The jet passed through a 1 mm skimmer into the detection chamber. The background pressure of the detection chamber was 2×10^{-10} mbar.

The vertically polarized alignment pulse was stretched to about 60 fs with a 15 mm quartz block, since best alignment is achieved if the pulse duration approximately matches the alignment time. Furthermore, we apertured the alignment beam to ensure that its focal volume was larger than that of the ionization beam. Finally, we worked at the highest possible intensity that creates a low ionization background ($< 1\%$) compared to the ionization pulses.

Knowing the intensity of the ionization pulse, we deduced the pump intensity to be $(2.5 \pm 1) \times 10^{13}$ W/cm² from the known intensity dependence of argon ionization yields as well as theoretical molecular ionization rates for N₂. This value agrees well with an intensity estimate from pulse length, pulse energy, and focus area.

The horizontally polarized ionization pulse was neither clipped nor chirped. From the momentum distribution of the electrons during single ionization, we calculate its intensity to be $(1.2 \pm 0.2) \times 10^{14}$ W/cm² [15].

We used cold target recoil ion momentum spectroscopy (COLTRIMS) to determine the momenta of electrons and ions [16,17]. They were directed via a uniform electric field of 5.6 V/cm and a magnetic field of 11.0 G towards two microchannel plates with delay line readout. From time of flight and position on the detectors, the three-dimensional momentum vector for each particle was obtained. For the electrons, the extraction length was 6.3 cm and the field-free length was 14.6 cm, whereas the corresponding lengths for the ions were 22.3 and 43.5 cm, respectively. We used the single ionization channel to measure the momentum resolution of the system. It is ± 0.18 a.u. along the spectrometer axis, which is parallel to the direction of the ionization laser, ± 0.37 a.u. along the propagation direction of the laser, and ± 1.37 a.u. along the jet direction, i.e., parallel to the direction of the alignment laser. From the last value, we deduce a translational jet temperature of 6 K. The rotational temperature can be estimated to be only slightly above this temperature [18]. The settings of electric and magnetic fields yielded a 4π solid angle for electrons up to 42 eV.

The electron count rate was 0.3 per shot and the ion rate 0.1. For single ionization, real coincidences are identified by momentum conservation between ion and electron. The fraction of false coincidences in which the registered ion and electron result from two different atoms, both ionized in the same shot, was measured to be 11% for double ionization.

The pump pulse creates a rotational wave packet in the nitrogen ensemble [11]. The wave packet quickly dephases, and rephases at half multiples of $\tau = 1/(2B_e) \approx 8$ ps. We use a delay of ≈ 4 ps where the angular distribution of the wave packet first peaks parallel to the alignment pulse polarization and then in a disk shape perpendicular to the alignment pulse shortly afterwards [19]. Throughout this Letter, though, we define perpendicular [Fig. 1(a)] and parallel [Fig. 1(b)] with respect to the ionization pulse polarization. The angle-dependent ionization rate thus minimizes at 3.93 ps when the ensemble is aligned perpendicular to the ionization pulse and maximizes at 4.30 ps when it is parallel [20,21]. Compared to unaligned molecules, we observe an increase and decrease, respectively, by more than 8% in the direct total count rate when switching the delay between 4.30 and 3.93 ps.

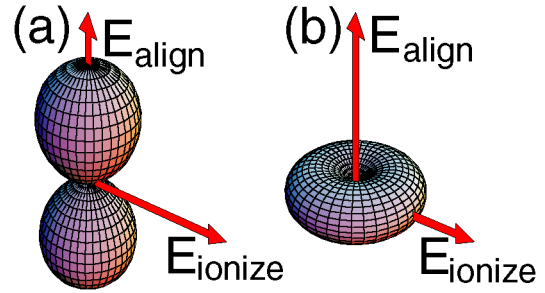


FIG. 1 (color online). Sketches of the alignment distribution for perpendicular (a) and parallel (b) molecules with respect to the alignment and ionization laser polarization. We observe an N_2^{2+}/Ne^+ ratio of 0.696 and 0.905 for geometry (a) and (b), respectively.

We verified that the first pulse produces aligned molecules with two independent methods: The first method is indirect and relies on the alignment-dependent ionization rate of molecules [21]. We introduced a neon/nitrogen gas mixture in the jet and used the neon ionization yield as a reference to find relative yields for the different time delays, i.e., alignments of the molecules. We observed 19% more N_2^{1+} ions and 30% more N_2^{2+} ions when the molecules are aligned parallel. This is in good agreement with published results [20].

Second, we detected all N^+ ions from the $N_2 \rightarrow N^+ + N$ channel if they were emitted within a small angle within the spectrometer axis, i.e., parallel to the ionizing field. This method provides a direct measure of alignment, as the fragments are emitted in direction of the molecular axis. For the two time delays, we normalize the number of N^+ fragments within 6° of the laser polarization to the total number of observed N^+ fragments. Per observed N^+ fragment, we find 24% more N^+ events within the direction of the ionizing laser for parallel molecules.

We now turn to measuring double ionization dynamics. We use COLTRIMS to obtain both electron momenta in coincidence. That is, we select only events where there is one N_2^{2+} ion and one electron and calculate the momentum of the second electron by using momentum conservation. We discriminated against the N^+ fragments by their large momentum, since they result from the break-up of N_2^{n+} molecular ions. The N_2^{2+} ions thus form a narrow peak on a broad background of N^+ .

For the double ionization channel, $N_2 \rightarrow N_2^{2+} + 2e$, we confirmed that nonsequential double ionization is caused by recollision by observing that the N_2^{2+}/N_2^{1+} yield decreases strongly with increasing ellipticity of the ionizing pulse. We note that at our ionization pulse intensity, the maximum return energy of the rescattered electron at the core in our experiment is $\sim (28 \pm 3.7)$ eV, which is approximately the field-free ionization potential of the N_2^+ ion of 29.6 eV. Thus, we work at the classical threshold of double ionization, and must consider the influence of the

laser field on the instantaneous ionization potential of the parent ion.

Figure 2 shows the momentum correlation in the direction parallel to the probe (ionizing) laser field for molecules oriented parallel and perpendicular to the probe laser polarization. The horizontal axis represents the momentum component parallel to the laser field direction k_a^{\parallel} of one electron, the vertical axis the parallel momentum component k_b^{\parallel} of the other electron. Both electrons are most likely to be ejected with a similar momentum of $k_a^{\parallel} = k_b^{\parallel} \approx 0.5$ a.u. in the same direction for both alignments.

Assuming an electric field $E(t) = E_0 \cos \omega t$, we can infer the emission time t_{ion} of the electrons from the final momenta of the electrons [9,12]:

$$k_{a,b}^{\parallel} = 2\sqrt{U_p} \sin \omega t_{\text{ion}}, \quad (1)$$

with U_p being the ponderomotive energy of the electron in the oscillating laser field. This means that the electrons are most likely emitted at $\sim 30^\circ$ off the field maximum. This is in accordance with the observations made for atoms [9,12,22–24] and molecules [13,25]. Since our recollision electron energy is at threshold, the main contribution for double ionization will come from the maximum energy electrons, which are emitted around 17° and return around the zero crossing of the electric field at 270° [9]. Both electrons exchange energy and are subsequently emitted around 330° within one laser half cycle as the laser field lowers the barrier sufficiently. Moreover, we infer from [9] and further experiments on double ionization that (i) the electron momenta at the time of emission are small, such that the laser field almost entirely determines the final momenta of the electrons, and (ii) that the postcollision electron interaction is sufficiently small as not to make Eq. (1) invalid.

However, there are two qualitative differences between the electron momentum correlation maps in Fig. 2 for molecules that are aligned parallel and perpendicular to the ionizing laser field polarization. It is clear from this figure that there are more electrons in quadrants 2 and 4 when the molecules are perpendicular. In fact, for the same number of events, there are 22% more electrons in these quadrants for perpendicular molecules than for parallel molecules. Events in quadrants 2 and 4 originate from impact ionization followed by delayed tunnel ionization [12]. Thus, our experiment qualitatively shows that double ionization is slower for perpendicular molecules.

Figure 3 highlights this difference. To obtain Fig. 3(a), we normalize the plots of Fig. 2 to the same number of counts, subtract the parallel from the perpendicular spectrum, and set all negative values to zero. For Fig. 3(b), we proceed in the same manner, but subtract the perpendicular from the parallel spectrum. Again, the increase in events in quadrants 2 and 4 is clearly visible. This substantial change is statistically significant: Assuming Poisson statistics for

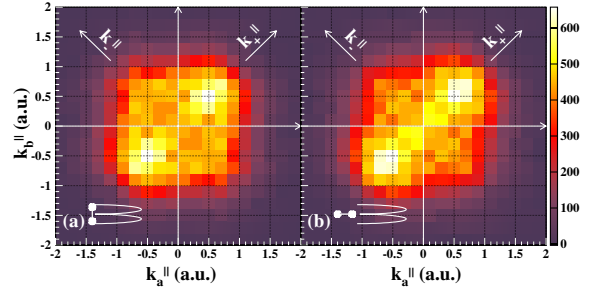


FIG. 2 (color online). Electron correlation for the double ionization of nitrogen at 1.2×10^{14} W/cm², 800 nm, 40 fs. (a) N₂ molecules oriented perpendicular, (b) parallel to the probe laser polarization (see sketches in the left bottom corner). Horizontal axis: momentum k_a^{\parallel} parallel to the probe field of electron a, vertical axis: parallel momentum k_b^{\parallel} of electron b. The bin size in both k_a^{\parallel} and k_b^{\parallel} direction has been chosen to match the momentum resolution in laser direction.

the events in Fig. 2, the error is given by \sqrt{N} where N is the number of events per bin in Fig. 2. For each bin in the difference spectra, Fig. 3, the error is thus $\sqrt{2N}$, where N is again the number of events in the corresponding bin in Fig. 2. The maximum value for $\sqrt{2N}$ in Fig. 2 is ~ 36 counts. This is below the number of events per bin in the difference spectra in any region where we observe significant changes in the correlation maps, which is in the range of 60–120 counts.

Second, Fig. 3 also shows that the average electron momentum is higher for parallel molecules. This can be seen from the on-diagonal maximum at ~ 0.9 a.u. in Fig. 3(b) and the on-diagonal maximum at ~ 0.4 a.u. in Fig. 3(a) [which corresponds to a minimum in Fig. 3(b)]. Fitting the positions of the peaks in Fig. 2, we find that the position of the maximum is shifted to larger momenta by (0.04 ± 0.025) a.u. for parallel molecules, resulting in a phase difference ωt of about $(2.8 \pm 1.7)^\circ$, corresponding of a time difference of $t = (21 \pm 13)$ attoseconds.

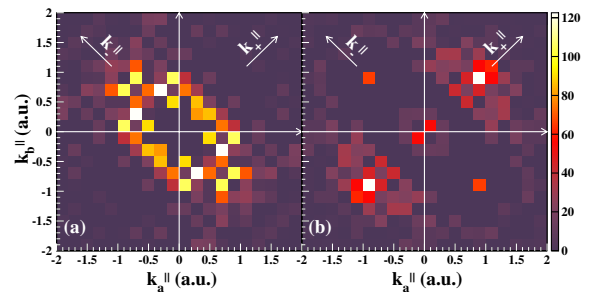


FIG. 3 (color online). Difference between the longitudinal electron correlation spectra shown in Fig. 2. Before taking their difference, the spectra have been normalized to their number of counts. For clarity, the graph has been split into two: (a) Perpendicular minus parallel, (b) parallel minus perpendicular, and all negative values have been set to zero.

Thus, our results show that the electrons are more likely to depart from the atom quickly (within the same half laser cycle) for parallel molecules in the same direction. The emission of one of the two electrons is more likely to be delayed for perpendicular molecules, and the electrons are rather emitted during different laser half cycles. Double ionization dynamics critically depends on the orientation of the scattering potential in which the two electrons interact, which is in contrast to the — to the best of our knowledge only—theoretical, classical analysis on double ionization of N_2 and O_2 [25] that predicts little or no effect of the electron correlation on alignment or molecular species.

Considering that the contrast of our parallel alignment is not perfect, the lower probability of nonsequential double ionization for perpendicular molecules is a significant effect. How is this possible? We propose that an aligned N_2 molecule represents a quasi-one-dimensional potential in which the electrons interact. When the molecule is parallel, recollision leads to doubly excited N_2 [26]. The recollision electron collides near zero field and shares its kinetic energy with the second electron while both are laterally confined by the potential, transferring their kinetic energy into potential energy. They are then ejected in rapid succession by the increasing field and their mutual repulsion [27]. In contrast, as the electrons interact during the recollision with a perpendicular molecule, they can evade each other laterally, which means that the energy and momentum exchange will be different in this geometry, translating into different emission times. When the field increases here, lowering the ionization barrier, their mutual interaction does not aid their joint departure.

Implicit in this interpretation is that the time dependence of the current density represented by the recolliding electron is insensitive to alignment. In other words, we assume that the kinetic energy spectrum of the electrons at the moment of recollision is unchanged. This is confirmed in a single ionization experiment of aligned N_2 [28] and by molecular tunnel ionization theory [21].

In conclusion, we have shown that during double ionization the electrons are ejected within different laser half cycles for perpendicular molecules, while they more often leave parallel molecules within the same laser cycle. This provides a strong constraint on possible mechanisms. Coulomb blockade of the electrons [29], as observed in the solid state, can be virtually ruled out. Instead, when the electrons are defined in a potential with 1 degree of freedom along the laser field polarization, they assist each other in becoming free. We propose to label this mechanism Coulomb-assisted ionization.

We thank T. Brabec, M. Meckel, O. Geßner, and M. Ivanov for valuable discussions. A.B. thanks the NRC

WES program, A.S. the Studienstiftung des deutschen Volkes, and D.Z. the Alexander von Humboldt-Stiftung for financial support.

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