

Molecular reorientation in intense femtosecond laser fields

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A femtosecond double-pulse experiment shows that molecular reorientation during multiple ionization is a very fast process for light molecules such as O₂ or N₂O using 60-fs laser pulses and variable time delays between ± 1 ps in the 10^{15} W/cm² intensity range. A unified two-dimensional model of molecular multiple ionization allows one to get the multicharged fragment trajectories and consequently discuss the speed of alignment of a molecule. The numerical results confirm that the molecular reorientation process is faster when the laser intensity increases, in good agreement with the experimental data.

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The first experimental reports of laser-induced multiple ionization of diatomic molecules have shown that the resulting multicharged fragments are preferentially produced along the laser polarization direction in the case of linearly polarized laser light [1–3]. Two explanations are then possible for this effect. The first one is based on a more efficient multiple ionization for molecules initially aligned along the laser electric field, while the second scenario involves a molecular reorientation due to the strong polarizability induced by the intense laser field. It has to be stressed that these two cases do not exclude each other, since reorientation can take place even in the case of a more efficient multiple ionization for molecules initially aligned with the laser polarization direction. The molecular reorientation was shown unambiguously for the CO molecule submitted to a 30-ps 1064-nm linearly polarized laser pulse in the 10^{14} – 10^{15} W/cm² intensity range, using a pump-probe laser excitation analysis [4]. However, heavier molecules such as I₂ in intense femtosecond laser fields are not reoriented, but exhibit a strong enhancement of the ionization rates when their internuclear axis is parallel to the laser electric field [5,6].

This paper is aimed at showing that molecular reorientation is a very fast process using a 60-fs pulse duration laser and a pump-probe diagnostic at $\lambda = 800$ nm for light molecules such as O₂ and N₂O in the 10^{15} W/cm² laser intensity range. Although the fragments emission patterns have been studied quite extensively [5,7–11], several questions remain open about the speed of molecular reorientation and the role of multiple ionization. For instance, the order of magnitude of the rotation time can be calculated using a simple model based on the neutral-molecule parallel and transverse polarizabilities [12]. At a laser intensity of 10^{15} W/cm² used typically in multiple ionization experiments, the rotation time is several hundreds of femtoseconds for neutral O₂ and N₂O molecules. In consequence, the laser-molecule coupling leading to the different molecular multiple ionization thresholds has to be taken into account. However, a full quantum time-dependent calculation remains very difficult due to the multitude of involved molecular states before reaching a particular dissociation channel. In this paper, we propose an alternative model based on a two-dimensional Thomas-Fermi approach [13]. This nonperturbative model is able to

reproduce the reorientation temporal behavior taking into account the molecular axis angle-dependent ionization of molecules in intense laser fields.

The experiments were performed at the Saclay LUCA laser facility (LUCA, Laser Ultra-Court Accordable). A 60-fs laser pulse is split into two pulses in a Michelson-like optical setup allowing one to get collinear output pulses with variable polarization directions and identical spatial properties. The laser-molecule interaction takes place in an ultrahigh vacuum chamber with a base pressure of 3×10^{-10} Torr, where the laser beam is focused by an on-axis parabolic mirror with focal length $F = 75$ mm. The laser spot and, in consequence, the peak laser intensity are estimated from the laser beam diameter (10 mm) and from the $M^2 = 4$ factor measurement [14]. This optical parameter characterizes the difference of our laser beam with an ideal Gaussian laser beam in which case $M^2 = 1$. The multicharged ion detection is performed using a Wiley-McLaren-type ion spectrometer, with a 780-mm-long time-of-flight tube [15,16]. Using a moderate collection electric voltage U_c in the interaction volume, only ions emitted along the spectrometer axis are detected. For an ion with initial energy E_0 and charge state Z , the maximum angle between the ion vector momentum and the spectrometer axis is given by $4.6(ZeU_c/E_0)^{1/2}$ deg, where e is the elementary charge.

The aim of the experiment is to probe the molecular population that remains after the Coulomb explosion induced by a linearly polarized laser pulse. In the case of no molecular reorientation during multiple ionization, a significant molecular population should be left in the perpendicular plane of the laser-polarization direction. This statement is consistent with the observed angular distributions that are peaked along the polarization direction [5,7–11]. The principle of the experiment is as follows. The two pulse polarization directions are set perpendicular to each other. For short, these pulses are called, respectively, the *parallel* pulse and the *perpendicular* pulse, where *parallel* and *perpendicular* indicate the pulse polarization directions with respect to the spectrometer axis. By convention, the time delay is negative when the parallel pulse arrives first and is varied from -1 ps to 1 ps. Following the spectrometer configuration, the detected multicharged fragment signals come mainly from

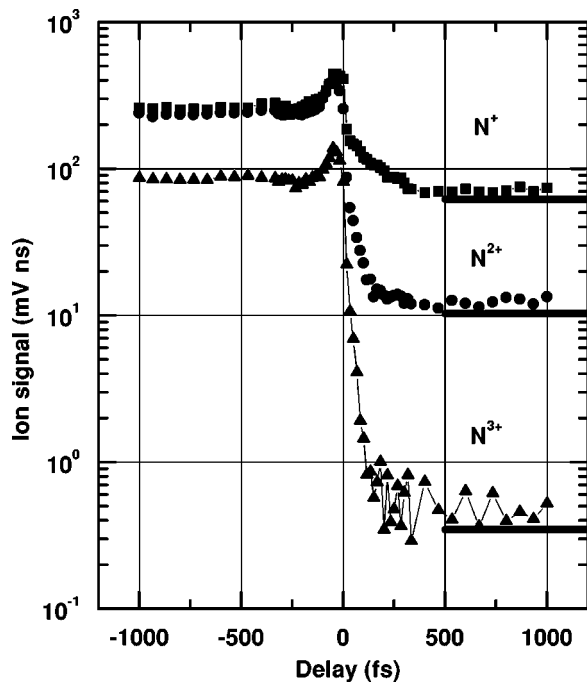


FIG. 1. Pump-probe experiment performed with the N_2O molecule at $p(N_2O) = 6 \times 10^{-8}$ Torr. The delay is negative when the parallel pulse arrives first. The laser intensities are 10^{15} W/cm 2 and 2×10^{15} W/cm 2 for, respectively, parallel and perpendicular pulses. The N^{Z+} ($Z = 1, 2, 3$) ion signals come from the terminal nitrogen atom of N_2O , and takes into account all the fragmentation channels that give rise to these ion species (see text). At positive delays, the horizontal lines represent the N^{Z+} ion signals recorded with the single perpendicular pulse using the same experimental conditions.

the parallel pulse, while the signals coming from the perpendicular pulse remain very weak. More precisely, the signal ratios between parallel and perpendicular excitations depend on the fragment angular distributions and spectrometer acceptance angle. The most interesting situation corresponds to positive delays when the perpendicular pulse arrives first. In the case of no molecular reorientation, the parallel pulse should find a significant molecular population that has not been multi-ionized, and in consequence should produce significant fragment signals. In the opposite case, a strong reduction of the fragment signals is expected. Finally, possible multiple ionization saturation processes are not relevant to this experimental analysis since fragment angular distributions of light molecules get narrower as the laser intensity is increased [5,10].

Figure 1 represents the N^{Z+} ($Z = 1, 2, 3$) ion signals behavior as functions of the time delay between the parallel and perpendicular pulses. These ion signals come from the terminal nitrogen atom N_t of $N_2O \equiv N_t N_c O$, where N_c represents the central nitrogen atom. The origin of these ions within a particular fragmentation channel $N_t^{Z+} + N_c^{Z'+} + O^{Z''+}$ is not taken into account, due to the large number of decay channels associated with a particular nitrogen multi-charged ion N_t^{Z+} . At negative delays, the parallel pulse arrives first in the interaction volume and produces significant fragment signals as is expected from single-pulse experi-

ments. At positive delays when the perpendicular pulse arrives first, a strong reduction of the fragment signals is observed. Indeed, the horizontal lines indicate the N^{Z+} signals produced by the single perpendicular pulse. Therefore, Fig. 1 shows that the late arrival of the parallel pulse has a very weak detectable effect. This observation shows that most of the molecular population has been reoriented by the perpendicular pulse. Indeed, if this pulse had ionized only molecules aligned along its polarization direction, there should remain a large population of molecules aligned along the parallel-pulse electric field. In this case, the ion signals should be comparable to ion signals detected at negative delays.

The time overlap between -100 fs and 100 fs between both pulses shows the combined contribution of both polarizations. As the negative time delay approaches zero, the molecular multi-ionization is still reoriented by the parallel pulse, but the perpendicular pulse tends to increase the overall multi-ionization efficiency. This process is maximum at negative delays of around -30 fs. This delay is not zero because the peak intensity of the perpendicular pulse is larger than the peak intensity of the parallel pulse by a factor of 2. Indeed as soon as the time-dependent intensity of the perpendicular pulse is higher than the peak intensity of the parallel pulse, the reorientation process is more effective for the perpendicular pulse. The value of -30 fs is consistent with the 60-fs full width at half maximum intensity of the initial laser pulse. After this time delay, the ion signals decrease rapidly due to the reorientation process induced by the perpendicular pulse. The contrast between ion signals at negative and positive delays increases noticeably as the fragment charge states get higher. This observation is in good agreement with the corresponding observed angular distributions, which are narrower as more and more electrons are removed from the molecule [5,7–11]. Accordingly, higher laser intensities within the interaction focal volume favor higher ionization stages and higher reorientation efficiencies.

The influence of the laser intensity is better illustrated for a diatomic molecule such as O_2 , since it is easier to isolate a single fragmentation channel within the time-of-flight spectrum. Figures 2(a) and 2(b) represent the O^{3+} ion signals coming from the $O^{3+} + O^{2+}$ fragmentation channel. This channel is identified using the covariance mapping technique [17] in a separate single-pulse experiment. Besides the double-pulse signals, the ion signals produced by the single parallel and perpendicular pulses are presented by horizontal lines at, respectively, negative and positive delays. For the highest intensities in Fig. 2(a) at negative delays, the difference between the double-pulse and single-parallel-pulse signals comes from the further ionization of O^{2+} ions from the same $O^{3+} + O^{2+}$ fragmentation channel by the perpendicular pulse. The main difference between Figs. 2(a) and 2(b) occurs at positive delays. In the case of low intensities in Fig. 2(b), the double-pulse signal does not return to the signal produced by the single perpendicular pulse. This experimental feature indicates that there remain a small fraction of molecules that have not been reoriented by the perpendicular pulse. This additional information shows that reorientation is a dynamical effect that is favored by high laser intensities.

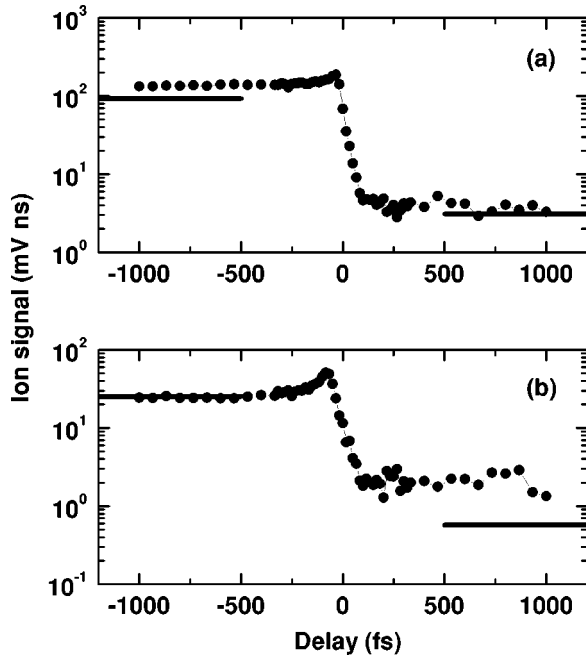


FIG. 2. Pump-probe experiment performed with the O_2 molecule at two different double-pulse laser intensities: (a) the laser intensities are 4×10^{15} W/cm 2 and 8×10^{15} W/cm 2 for, respectively, parallel and perpendicular pulses, (b) the laser intensities are 7×10^{14} W/cm 2 and 1.4×10^{15} W/cm 2 for, respectively, parallel and perpendicular pulses. The ion signal is recorded for the O^{3+} ion from the $O^{3+} + O^{2+}$ fragmentation channel. The delay is negative when the parallel pulse arrives first. At negative delays, the horizontal lines represent the O^{3+} ion signals recorded with the single parallel pulse. At positive delays, the horizontal lines represent the O^{3+} ion signals recorded with the single perpendicular pulse.

The main difficulty with calculations of reorientation processes comes from the fact that the model does have to take into account the laser-molecule coupling during the molecular rotation as well as the multiple ionization and subsequent Coulomb explosion. The two-dimensional Thomas-Fermi model is well suited for such a complicated molecular dynamics in strong fields, and has been presented elsewhere [13,18,19]. In brief, the molecular electrons are treated as an ideal, locally uniform, degenerate Fermi gas in the simplest approximation at zero temperature. The evolution of the electronic density $\rho(\vec{r},t)$ is calculated following the set of equations

$$\frac{\partial \rho(\vec{r},t)}{\partial t} + \vec{\nabla} \cdot [\rho(\vec{r},t)\vec{v}(\vec{r},t)] = 0,$$

$$\frac{\partial \vec{v}(\vec{r},t)}{\partial t} + [\vec{v}(\vec{r},t) \cdot \vec{\nabla}] \vec{v}(\vec{r},t) = - \frac{1}{\rho(\vec{r},t)} \vec{\nabla} P(\vec{r},t) + \vec{\nabla} \Phi(\vec{r},t), \quad (1)$$

where $\vec{v}(\vec{r},t)$ and $P(\vec{r},t)$ are, respectively, the velocity and pressure fields of the electron gas. The electric potential

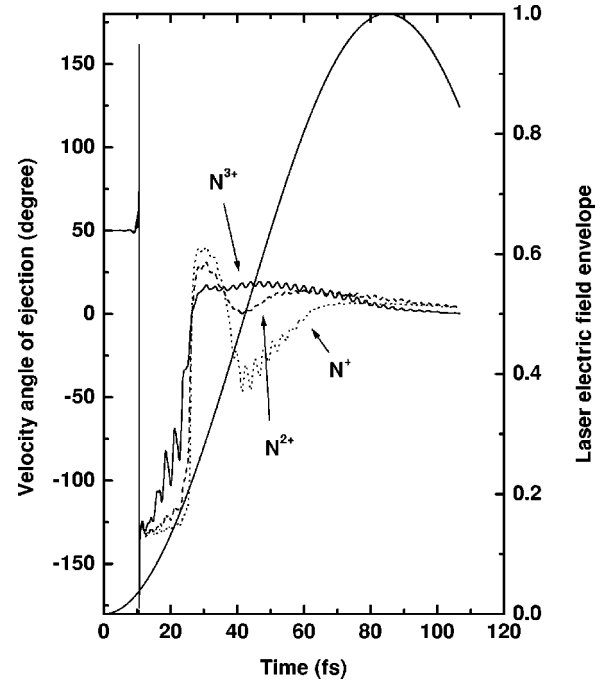


FIG. 3. Angle of the velocity (left vertical axis) of the terminal nitrogen ions as a function of time for initial orientation of N_2O molecular axis equal to 50° . The ions $N^{Z+} \equiv N_t^{Z+}$ ($Z=1,2,3$) belong to the $N_t^{Z+} + N_c^{Z+} + O^{Z+}$ fragmentation channel. The laser peak intensities are $I_1 = 5.6 \times 10^{15}$ W/cm 2 , $I_2 = 1.3 \times 10^{16}$ W/cm 2 , and $I_3 = 3.5 \times 10^{16}$ W/cm 2 for, respectively, $Z=1$, $Z=2$, and $Z=3$. The right vertical axis corresponds to the laser electric-field envelope, which is the unlabeled curve (see text).

$\Phi(\vec{r},t)$ is calculated as a sum of electric potentials originating in all the components of the system (electrons and nuclei) and in the laser field.

We have performed numerical studies for the N_2O molecule by solving the set of Eqs. (1) simultaneously with classical equations of motion for the nuclei (ions). The time dependence of the linearly polarized laser field $F(t)$ is introduced using a \sin^2 envelope according to $F(t) = F_0 \sin^2(\pi t/T_F) \sin(2\pi t/T_{Opt})$, where F_0 is the peak electric field and $T_{Opt} = 2.67$ fs is the optical period at the carrier wavelength $\lambda = 800$ nm. The field time duration T_F is set to $T_F = 169.7$ fs in order to reproduce the experimental pulse width $T_1 = T_F / (2\sqrt{2}) = 60$ fs, where T_1 is the full width at half maximum of the laser intensity time envelope. The maximum electric field value F_0 is chosen so that the molecule gets ionized in the rising edge of the pulse. The resulting Coulomb explosion is developed within the interval 25 to 50 fs before the laser peak field is reached at $t = T_F/2 = 85$ fs. Finally, the initial molecular orientation with respect to the laser field direction is equal to 50° .

The relevant physical quantities to the experiments are the angles of ejection of the fragments velocities, which are defined as $\arctan(v_y/v_x)$, where v_x and v_y are the two-dimensional velocities components, with the Ox axis parallel to the laser electric field. These angles are represented in Fig. 3 for the terminal nitrogen ions $N^{Z+} \equiv N_t^{Z+}$ ($Z=1,2,3$) that belong to the $N_t^{Z+} + N_c^{Z+} + O^{Z+}$ fragmentation channel. The

laser peak intensities are $I_1=5.6\times 10^{15}$ W/cm², $I_2=1.3\times 10^{16}$ W/cm², and $I_3=3.5\times 10^{16}$ W/cm² for, respectively, $Z=1$, $Z=2$, and $Z=3$. The laser electric-field envelope is also represented in Fig. 3 to show the reorientation of the overall molecular system during the laser pulse duration. At the beginning of the pulse, the angles are equal to 50° with a sudden change at 11 fs. This behavior is observed for the three ion classes N^+ , N^{2+} , and N^{3+} , and is due to the molecular vibration along the molecular axis. The vibration results in a change of the velocity angle by 180° from 50° to -130° . A second sharp reversal of the angles occurs at 26 fs, although an increasing departure from vibrational motion can be seen for the N^{2+} and N^{3+} ions. After 26 fs, the angle oscillations are damped to end up with final velocities aligned along the laser-polarization direction. The overall behavior of the curves in Fig. 3 shows that the leading edge of the pulse is already very efficient in getting molecular fragments aligned. The reorientation efficiency increases with the fragment's final charge state, i.e., the laser peak intensity. Indeed, the damping of the angle oscillations appears to be more and more pronounced as the laser intensity is increased. During Coulomb explosion, the intense laser field enforces the oscillations of the electronic cloud along the polarization vector. This oscillating charge influences the motion of the exploding ions. The attractive Coulomb force drags the ions towards the direction of the electric field. The deflection of the exploding ions trajectories is then the mechanism of the alignment of the fragments supported by the model. Higher laser peak intensities trigger electronic oscillations sooner in the laser pulse, and in consequence are more efficient for the overall reorientation of the molecular

system. The peak laser intensities used in the calculations are higher than the experimental values because the calculation's code produces only the $N^{Z+}+N^{Z+}+O^{Z+}$ fragmentation channel while the experimental N^{Z+} ion signals come from this channel as well as channels with lower charge state companion fragments $N^{(Z-1)+}$ and/or $O^{(Z-1)+}$. In addition, the two-dimensional approach of the model might be another source for this difference. A more thorough comparison would require the introduction of the isotropic initial molecular distribution as well as the laser intensity distribution within the focal volume using a full three-dimensional model. Such a work is underway in order to reproduce the observed angular distributions.

In conclusion, this paper shows that molecular reorientation in strong laser fields is an ultrafast process that can be as short as 60 fs for light molecules. The hydrodynamic model provides a meaningful description of molecular Coulomb explosion and supports the observed very fast reorientation processes. The reorientation speed increases with the laser intensity, in agreement with the experimental data. This effect can be interpreted as a generalized intensity-dependent electronic polarizability in the course of molecular multiple ionization.

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