Optical Imaging of Coherent Molecular Rotors

Jérémy Bert, Emilien Prost, Ilia Tutunnikov, Pierre Béjot, Edouard Hertz, Franck Billard, Bruno Lavorel, Uri Steinitz, Ilya Sh. Averbukh,* and Olivier Faucher*

Short laser pulses are widely used for controlling molecular rotational degrees of freedom and inducing molecular alignment, orientation, unidirectional rotation, and other types of coherent rotational motion. To follow the ultrafast rotational dynamics in real time, several techniques for producing molecular movies have been proposed based on the Coulomb explosion of rotating molecules, or recovering molecular orientation from the angular distribution of high harmonics. The present work offers and demonstrates a novel nondestructive optical method for direct visualization and recording of movies of coherent rotational dynamics in a molecular gas. The technique is based on imaging the time-dependent polarization dynamics of a probe light propagating through a gas of coherently rotating molecules. The probe pulse continues through a radial polarizer, and is then recorded by a camera. The technique is illustrated by implementing it with two examples of time-resolved rotational dynamics: alignment–antialignment cycles in a molecular gas excited by a single linearly polarized laser pulse, and unidirectional molecular rotation induced by a pulse with twisted polarization. This method may open new avenues in studies on fast chemical transformation phenomena and ultrafast molecular dynamics caused by strong laser fields of various complexities.

1. Introduction

Resolving the motion of molecular nuclei during chemical transformations, or when under the influence of ultrafast external fields, is of extreme importance in chemistry and physics. The ultimate goal is recording the so-called “molecular movies” unveiling the corresponding molecular dynamics in “real time.” Filming molecules involves many different time scales, from short ones relevant to the electronic dynamics, to the longest ones corresponding to molecular rotation. Recent advances in high-intensity electron and X-ray pulsed sources made it possible to directly observe atomic motions and electron dynamics happening on intramolecular scale. As it is easier to follow and control the slow motion, molecular rotational dynamics became a testing ground for developing new methods for molecular “cinematography,” and various approaches for quantum control of molecular dynamics. In particular, detailed “movies” were recorded for imaging unidirectional rotation of linear molecules subject to strong laser pulses with crossed linear polarizations, and extremely rich rotational dynamics of molecules undergoing field-free alignment–antialignment cycles. Detection methods for probing the angular localization of molecules resulting from strong laser field interactions can be divided into two categories. The first one is based on fragmentation of the molecules, while the second one relies on optical detection. In the first category, short and intense resonant laser pulses are used for breaking molecular bonds through a resonant dissociation process followed by ionization of the fragments or through Coulomb explosion. In the case of a linear molecule, molecular fragments are assumed to recoil along the line defined by the molecular axis at the moment of molecular ionization. This allows retrieving the orientation of the molecule from velocity map imaging (VMI) of the photofragments. Because the technique is sensitive to the charge trajectories projected on the plane of a detector, the VMI provides a 2D information about the angular distribution of the molecules. In contrast, a full 3D information can be accessed by cold target recoil ion momentum spectroscopy (COLTRIMS), where both electron and ion momenta are imaged in coincidence. Methods based on charge momentum imaging provide a wealth of information about the molecular dynamics and therefore constitute powerful tools for their full characterization. However, such techniques require sophisticated apparatus, long acquisition times, and meticulous data postprocessing, and are applicable only to the rarefied and cold molecular gases. In contrast, optical techniques do not require highly dedicated apparatus, can be easily implemented, and are compatible with a wide range of gas pressures and temperatures. So far, optical detection schemes have been limited to 1D measurements of the ensemble.
averaged quantities, like orientation factor, $\langle \cos \theta \rangle^{[14]}$ and alignment factor, $\langle \cos^2 \theta \rangle^{[15]}$ where $\theta$ is the Euler angle between the molecule-fixed and a space-fixed axes. They have also been used to measure higher-order moments $\langle \cos^n \theta \rangle$, with $n > 2$, of the angular distribution by using high-harmonic generation.[16] Recently, angle-resolved high-order-harmonic spectroscopy was used for generation of a molecular “rotational movie” with the help of iterative machine learning procedure.[17]

The present work introduces a new optical technique for direct time-resolved imaging of coherent molecular rotation in the gas phase. It relies on the use of a vortex plate which can be used to generate beams with a nonuniform state of polarization, known as vector beams. The latter, and in particular those with radial or azimuthal polarizations, have been the subjects of considerable interest[18] largely motivated by their unique focusing properties.[19,20] Radial and azimuthal vector beams find applications, for instance, in laser machining,[21,22] plasmonics,[23] optical trapping,[24] and tabletop extreme ultraviolet coherent sources.[25] Here, the vortex plate is used to achieve a direct visualization of the angular localization of molecules in space. The technique is applied to observing ultrafast temporal alternation between molecular alignment and antialignment in a molecular gas excited by a short nonresonant linearly polarized pulse, as well as to monitoring coherent unidirectional rotation of molecules in the case of excitation by a pulse with twisted polarization.

2. Concept

2.1. Aligned Molecules

Laser-induced molecular alignment results from the anisotropic interaction between the electric field of a strong laser pulse and the induced molecular dipole. The concept was introduced in the early 90s[26,27] and was initially investigated in the adiabatic regime, using pulse durations much longer that the rotational period $T$ of the molecule. In general, long pulses allow achieving high degree of alignment, even at moderate temperatures. However, a drawback of the adiabatic interaction is that the alignment exists only in the presence of a strong external field, which may complicate the interpretation of the experimental results and also limits the potential applications. As an alternative, impulsive alignment was proposed[28,29] and developed, allowing preparation of aligned molecular samples under field-free conditions.[30] Although our imaging technique is in principle applicable to both the adiabatic and nonadiabatic cases, the present work focuses on the latter regime, that represents a widely used common approach for aligning molecules with lasers (more details about laser-induced molecular alignment can be found in refs. [31–33]).

Figure 1a depicts a calculated alignment factor of CO$_2$ molecules at room temperature kicked by a nonresonant short laser pulse linearly polarized along $z$ axis, which is chosen as the quantization axis for the numerical simulation. The alignment process is studied by solving the Liouville–von Neumann equation for molecules driven by a Gaussian laser pulse with peak intensity of 20 TW cm$^{-2}$ and duration of 100 fs at full width half maximum (FWHM), which is applied at $t = 0$. The theoretical model, described in ref. [34], allows computing molecular rotational dynamics for any polarization of the excitation pulse. The values of the alignment factor $\langle \cos^2 \theta \rangle > 1/3$ signify the preferential alignment of the molecular axis along the polarization direction of the field. A state in which the molecules are preferentially localized in a plane perpendicular to the field is called antialignment, and it is characterized by $\langle \cos^2 \theta \rangle < 1/3$. During the field-free evolution of the quantum system after the turnoff of the aligning field ($t > 0.1$ ps), the anisotopic angular molecular distribution alternates between alignment and antialignment within each rotational revival recurring with a period given by $T/4$ (see, e.g., the corresponding angular distributions of molecules depicted around the third fractional revival at $3 T/4$). Between the revivals, a residual permanent alignment is manifested by a value of $\langle \cos^2 \theta \rangle$ standing slightly above its isotropic value of 1/3.[35]

![Figure 1](https://www.advancedsciencenews.com/doi/10.1002/1432-0242.00100011)
2.2. Spinning Molecules

If instead of exciting the molecules by a linearly polarized laser pulse, a field with twisted linear polarization\[^{[36]}\] is applied to the system, then a unidirectional rotation of the molecules can be induced in the plane of polarization, according to the mechanism described in refs. \[^{[37,38]}\]. The molecular angular momentum gets oriented along the direction of the field propagation, and the sense of rotation can be controlled by adjusting the phase of the shaped pulse.\[^{[34]}\] Other field configurations leading to similar effect include a chiral trains of pulses,\[^{[39]}\] a pair of delayed polarization-crossed pulses,\[^{[37,38,40,41]}\] and an optical centrifuge.\[^{[12–44]}\]

Field-free evolution of the molecular ensemble excited by a twisted field can be captured by measuring the value of \(\langle \cos^2 \phi \rangle\), where \(\phi\) is the azimuthal Euler angle (relative to the \(x\) axis) in the \(xy\) plane in which the polarization of the field is confined. Notice that unlike the previous result depicted in Figure 1a, here, the quantization axis \(z\) is chosen along the propagation axis of the field. This observable allows to estimate when on average, molecules are directed along the \(x\) axis (\(\langle \cos^2 \phi \rangle > 1/2\)), or the \(y\) axis (\(\langle \cos^2 \phi \rangle < 1/2\)), while the isotropic distribution corresponds to \(\langle \cos^2 \phi \rangle = 1/2\). Figure 1b shows the simulated result for CO\(_2\) after the molecules have been excited by a sequence of two orthogonally polarized pulses of same duration (100 fs) and peak intensity (20 TW cm\(^{-2}\)) separated by 145 fs and phase synchronized. One can see that during the revivals, the molecular behavior resembles the rotation of an aircraft propeller. This fast coherent molecular rotation has been detected in the past with the help of rotational Doppler effect resulting from the exchange of energy and angular momentum between the spinning molecules and a circularly polarized probe optical field.\[^{[45,46]}\]

3. Experimental Results

3.1. Experimental Method

From the point of view of optics, a gas of anisotropically distributed molecules is a birefringent medium, characterized by its principal optical axes. Here, we consider a gas of linear molecules that has been transiently aligned or spun by a nonresonant laser pulse (pump). Note that although aligned molecules macroscopically act as a uniaxial birefringent system, spinning molecules behave as a biaxial optical system with three distinct refractive indices due to the lack of axial symmetry in the angular distributions of the molecules. For optical imaging, the sample is probed by a time delayed circularly polarized short pulse (probe). During the probe propagation, the medium is assumed to be stationary. The probe field is decomposed along the two principal axes of the anisotropic medium. Each polarization component propagates through the medium under a different refractive index resulting in accumulation of relative phase between them. After passing the medium, the probe becomes elliptically polarized with the major axis of the ellipse oriented at \(\pm \pi/4\) to the slow axis of the medium (the sign depends on the sense of circular polarization).

In order to locate the orientation of the ellipse, the probe field is directed to a 2D polarization analyzer combining a vortex plate and a linear polarizer. The zero-order vortex \(m = 1\) retarder consists of half-wave plates whose local orientations \(\chi\) vary continuously with the azimuthal angle \(\Phi\) of the plate according to the law \(\chi = m\Phi/2\) (Figure 2).

The principle of action of the 2D polarization analyzer is presented in Figure 2. As depicted in Figures 2a,b, the vortex plate converts a linear horizontal polarization into a radial polarization, and inversely, a vertical linear polarization into an azimuthal polarization. The direction of polarization of the incident light is retrieved by imaging the intensity of the transmitted beam on a charge-coupled device (CCD) camera. To this aim, a linear polarizer oriented parallel to the symmetry axis of the vortex plate (horizontal direction in Figure 2) is inserted between the vortex plate and the CCD camera. As shown, in both cases the symmetry axis of the intensity pattern rendered on the CCD camera is parallel to the major axis of the polarization ellipse of the incident light.

When the molecules are randomly oriented, the circular polarization of the probe passing through the vortex plate results in an isotropic intensity distribution on the CCD camera. However, when the isotropy of the medium is broken, e.g., during a molecular rotational revival, the probe light becomes elliptically polarized. This situation is illustrated in Figure 2c, where we assume that before the vortex plate, in each point in the plane, the major axis of the polarization ellipse is oriented at \(\pm \pi/4\) with respect to the axis of molecular alignment. After the vortex plate, all ellipses are differently tilted depending on the angular position across the beam profile, except at \(-\pi/4\) (\(\pi/4\)) where the major...
Figure 3. a) Experimental setup for producing and imaging coherent molecular rotation. The left side of the figure is related to the preparation of a coherent excitation of gas phase molecules by a polarization shaped femtosecond laser pulse (pump). The right side describes the imaging device used to record the spatial orientation of the molecules detected by a probe pulse. (BS, beam splitter; HWP, half-wave plate; P, polarizer; BBO, type-I phase-matching $\beta$-barium borate crystal; MOWP, multiple-order wave plate; BC, Berek compensator; DM, dichroic mirror; L, lens; VP, vortex plate; F, UV bandpass filter; BD, beam dump; ND, neutral density; F, filter.) b) The polarization shaper consisting in a MOWP and a BC generates two partially time overlapped, cross-polarized in phase (or out of phase) short laser pulses resulting in a field with a twisted linear polarization.\[36\]

3.2. Experimental Setup

The experiment is based on a femtosecond Ti:sapphire chirped-pulse amplifier producing pulses of 100 fs duration (FWHM) with a repetition rate of 1 kHz and a maximum energy of 3 mJ. The overall experimental setup is shown in Figure 3a. The polarization of the pump pulse ($\omega$) is shaped by a multiple-order wave plate (MOWP) combined with a Berek compensator (BC$_1$). By changing the polarization direction of the incident field, it is possible to generate at the exit of the shaper\[36\] a pulse exhibiting a fixed linear polarization or a twisted linear polarization as illustrated in Figure 3b. The former is used to impulsively align the molecules, whereas the latter produces transiently spinning molecules, as previously described in Section 2. The probe beam is frequency doubled ($2\omega$) with a BBO crystal and then overlapped with the pump beam. The Berek compensator (BC$_2$), placed before the dichroic mirror (DM$_2$), has its retardance adjusted in order to ensure that the probe pulse is circularly polarized after reflection by DM$_2$. A motorized retroreflector is introduced in the optical path of the pump beam for controlling the optical delay $\tau$ between the pump and probe beam. Both beams are focused with a lens ($L_1$) in a static cell filled with room temperature CO$_2$ gas. A second lens ($L_2$) is inserted before the vortex plate in order to collimate the probe beam. To avoid the pump light reaching the detector, two dichroic mirrors and a filter (F) are placed before the CCD camera.

3.3. Results and Discussions

We start with the results related to molecular alignment imaging. The snapshots presented in Figure 4 have been recorded in CO$_2$ gas under a pressure of 1 bar when the pump–probe delay is tuned over the third alignment revival 3$T_r$/4 discussed in Figure 1a. The movie of the whole sequence can be found...
in the Supporting Information. The aligning pulse (pump) is horizontally polarized in the plane of the camera. Recall that depending on the handedness of the probe’s polarization, the present technique produces a $\pm \pi/4$ angle between the molecules and the major axis of the polarization ellipse of the probe field imaged on the detector. Therefore, for convenience, all images presented in this work have been rotated by $\pi/4$, so that the symmetry axis of the images coincides with the molecular axis.

Figure 5 shows two types of images, one with an elongated intensity distribution along the horizontal axis, i.e., parallel to the pump polarization, and another one elongated along the vertical axis. Note that the vortex plate transforms the incident Gaussian probe beam TEM$_{00}$ profile into a Laguerre–Gaussian beam LG$_{01}$, which explains the attenuation of the signal observed near the singularity at the center of each snapshot. In the first group of images, the molecules are aligned along the polarization direction of the pump laser field, whereas in the second group, they are delocalized (antialigned) in the plane perpendicular to the pump. The maximal degree of alignment is observed around $\tau = 32.07$ ps, which is in good agreement with the simulation presented in Figure 1a, predicting the moment of maximal alignment around $t_0 = 32.06$ ps for the $3T_e/4$ revival. The antialignment reaches its maximum value for $\tau = 31.68$ and 32.45 ps, which is also consistent with the calculated values of $t_0$ and $t_c$. The results of Figure 4 provide a direct visualization of the complex spatiotemporal dynamics of the molecular alignment process. In the course of each revival, molecules toggle between alignment and antialignment; two different coherent states of the molecular rotor.

The images presented in Figure 5 have been recorded when the molecules are spun by a pulse exhibiting a twisted linear polarization. The latter executes a clockwise rotation corresponding to a quarter turn, starting from the vertical direction and ending horizontally. It triggers a unidirectional rotation of the molecules, which is here captured during the third revival of CO$_2$. Note that similar dynamics can be observed for other revivals. The accuracy of the imaging technique can be attested by comparing the measurements to the simulation of $(\cos^2 \phi)$. In particular, consider the images recorded around the specific times $t_3$ and $t_4$ introduced in Figure 1b. The local minimum $t_3$ of the function $(\cos^2 \phi)$ implies a preferential alignment of the molecules along the $y$ axis, which is well observed if one inspects the image recorded at the corresponding time $\tau = 32.05$ ps. In contrast, the local maximum at $\tau = t_4$ suggests that the molecular axes are directed along the $x$ axis, which is also confirmed by the images obtained at $\tau = 32.29$ ps. The coherent rotation of the molecules, which, it should be noted, cannot be inferred from $(\cos^2 \phi)$, is noticeable when one observes the successive images recorded between $\tau = 32.05$ and 32.29 ps. The clockwise sense of rotation is also clearly evidenced by the corresponding movie provided in the Supporting Information.

Although the technique is tested with linear molecules, it can be applied to any other molecular systems, including asymmetric top molecules and more generally any birefringent uniaxial and biaxial systems. The reason is that the polarization of the field that drives the molecular rotation is confined to the plane of polarization of the probe. In this case, the propagation axis of the fields is, independently of the nature of the microscopic system, a principal axis of the permittivity tensor and therefore the two neutral axes lying in the plane of observation coincide with the two other principal axes. For a linear molecule, in which the slow axis corresponds to the molecular axis (symmetry axis), i.e., the axis of largest polarizability, the present imaging technique allows the observation of the molecular bond alignment axis. For all asymmetric top molecules except chiral molecules, the principal axes coincide with the axes of inertia of the system. Therefore, as for linear, or more generally any symmetric top molecules, the technique can still be used to observe the molecular axis that is aligned or spun in the plane of observation. Only in the case of chiral species, a precise knowledge of the molecular system allows retrieving the orientation of the aligned molecular bond with respect to the neutral axis of the medium.

The next section describes how quantitative information about the instantaneous orientation and angular velocity of the slow axis of molecular gas can be extracted from the image sequence presented in Figures 4 and 5.

4. Data Analysis and Numerical Simulations

The birefringence and orientation of the molecular sample can be fully assessed by analyzing the angular dependence of the images $M(R, \Phi)$, where $R$ and $\Phi$ are the radial and azimuthal coordinates of the camera, respectively. The angular dependence $S(\Phi)$ at each delay $\tau$ is obtained from the corresponding image by summing the signal over $R$. Figure 6a,b depicts the delay-dependent angular signal measured with a pump pulse exhibiting a fixed and twisted linear polarization, respectively. Assuming a cylindrically symmetric beam intensity pattern, one can show using the Jones
matrix formalism that the angular dependence of the signal intensity $S(\Phi)$ can be written

$$S(\Phi) \propto 1 + \sin \psi \sin[2(\Phi - \phi_s)]$$

(1)

where $\psi$ is the phase difference experienced by the probe field components along and perpendicular to the slow axis of the medium, i.e., the molecular axis in the present study, the latter making an angle $\phi_s$ with respect to the $x$ axis of the laboratory frame (Section 2.2). It is clear from Equation (1) that all the information about the molecular sample is embedded in the oscillating term. For instance, we can see that the signal is maximal for $\Phi = \phi_s + \pi/4 \mod \pi$, as described in Section 3.1, and that the amplitude of the oscillation is directly proportional to the dephasing $\psi$, which is related to the alignment of the molecules. For the data analysis, it is convenient to cast $S(\Phi)$ in the form

$$S(\Phi) \propto 1 + \frac{\sin \psi}{2} e^{-i[\pi/2 + 2\phi_s]} e^{i2\phi_s} + \frac{\sin \psi}{2} e^{i[\pi/2 + 2\phi_s]} e^{-i2\phi_s}$$

(2)

which is none other than the Fourier series of the signal defined as

$$S(\Phi) = \sum_{n=-\infty}^{\infty} S_n e^{-i\Phi}$$

(3)

where $S_n$ is the $n$th complex valued coefficient of the Fourier series. The information about the molecular sample ($\phi_s$ and $\sin \psi$) can be directly retrieved from the second Fourier coefficient $S_2$

$$|\sin \psi| \propto |S_2|$$

(4)

$$\phi_s = \frac{\text{arg}(S_2)}{2} - \pi/4 \mod \pi$$

Note that Equation (4) can also be used as a reasonably good approximation of $\sin \psi$ and $\phi_s$ in case the angular dependence of the experimental signal is affected by any imperfection of the detection setup, as for instance a residual ellipticity of the probe beam or a nonperfect cylindrical laser beam (Section S2.C, Supporting Information). For completing the analysis of the experimental data, we have also estimated the birefringence $\Delta n$.
between the neutral axes of the medium and the instantaneous angular velocity of the medium $\omega(t) = \partial_j \phi_j$. As described in Section S3 in the Supporting Information, this was achieved by fitting the shape of the second Fourier coefficient $S_2$ with the calculated birefringence $\Delta n$. $\Delta n$ and $\omega(t)$ are depicted in Figures 6a,b, respectively.

In order to test the accuracy of the optical imaging technique, we have computed the time-dependent angular distribution of the molecules, from which the birefringence $\Delta n$ of the medium and the average angular velocity $\omega$ of the molecular axes can be estimated. As shown in Figure 6, the calculated values are in very good agreement with the data extracted by analyzing the recorded images. The latter are also well reproduced by the theory if one compares the records of Figure 5 with the simulations of Figure 7 representing the angular distributions of the spinning molecules calculated at the same delays. Note that the last calculations have been performed by choosing the direction of propagation of the pulses as a quantization axis, meaning that the Euler angle $\phi$ and the angle of observation $\Phi$ are equivalent in this case.

5. Conclusion

In summary, we have developed an optical method for imaging quantum rotors that are angularly localized in space. To the best of our knowledge, it is the only all-optical imaging technique reported so far enabling the visualization of ultrafast molecular rotation. It relies on the spatially resolved detection of transient birefringence performed in a pump–probe configuration. The key element of the detection scheme is a vortex half-wave retarders plate that combined with a linear polarizer and a camera allows to capture the orientation of the molecules across the beam of a femtosecond laser pulse. To this aim, the latter is circularly polarized before its interaction with the molecular gas. The imager is successfully employed to locate the orientation of the molecules after they have been either aligned or spun by an extra short laser pulse. By tuning the delay between the two laser pulses, one can record, on a single shot basis, the ultrafast coherent rotation of a molecular ensemble. Besides the orientation of the medium, it is demonstrated that useful additional information, as the birefringence or average angular velocity of the aligned molecules in the medium, can be extracted by analyzing the recorded images. More generally, the present optical method locates the neutral axes of a birefringent medium and therefore is in principle applicable to any nonisotropic transparent medium including molecular superrotors, nanostructured, macroscopic objects, and metamaterials.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

J.B., E.P., and I.T. contributed equally to this work. This work was supported by the CNRS, the ERDF Operational Programme - Burgundy, the EIPHI Graduate School (Contract No. ANR-17-EURE-0002), the Associate (CNRS&Weizmann) International ImagiNano Laboratory, and the Israel Science Foundation (Grant No. 746/15). Calculations were performed using HPC resources from DNUM-CCUB (Université de Bourgogne). I.S.A. acknowledges support as the Patricia Elman Bildner Professorial Chair. This research was made possible in part by the historic generosity of the Harold Perlman Family.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

optical imaging, spinning molecules, ultrafast birefringence, vortex half-wave retarder

Received: October 11, 2019
Revised: February 14, 2020
Published online: March 11, 2020
[16] S. J. Weber, M. Oppermann, J. P. Marangos, *Phys. Rev. Lett.* 2013, 111, 263601.
[17] Y. He, L. He, P. Lan, B. Wang, L. Li, X. Zhu, W. Cao, P. Lu, *Phys. Rev. A* 2019, 99, 053419.
[18] Q. Zhan, *Adv. Opt. Photonics* 2009, 1, 1.
[19] R. Dorn, S. Quabis, G. Leuchs, *Phys. Rev. Lett.* 2003, 91, 233901.
[20] L. Novotny, M. R. Beversluis, K. S. Youngworth, T. G. Brown, *Phys. Rev. Lett.* 2001, 86, 5251.
[21] V. G. Niziev, A. V. Nesterov, *J. Phys. D* 1999, 32, 1455.
[22] M. Meier, V. Romano, T. Feurer, *Appl. Phys. A* 2007, 86, 329.
[23] Q. Zhan, *Opt. Express* 2004, 12, 3377.
[24] C. Hernández-García, A. Turpin, J. San Román, A. Picón, R. Drevinskas, A. Cerkauskaite, P. G. Kazansky, C. G. Durfee, I. N. J. Sola, *Optica* 2017, 4, 520.
[25] D. Normand, L. A. Lompre, C. Cornaggia, *J. Phys. B* 1992, 25, 497.
[26] B. Friedrich, D. Herschbach, *Phys. Rev. Lett.* 1995, 74, 4623.
[27] T. Seideman, *Phys. Rev. Lett.* 1999, 83, 4971.
[28] J. Ortigoso, M. Rodríguez, M. Gupta, B. Friedrich, *J. Chem. Phys.* 1999, 110, 3870.
[29] F. Rosca-Pruna, M. J. J. Vrakking, *Phys. Rev. Lett.* 2001, 87, 153902.
[30] H. Stapelfeldt, T. Seideman, *Rev. Mod. Phys.* 2003, 75, 543.
[31] Y. Ohshima, H. Hasegawa, *Int. Rev. Phys. Chem.* 2010, 29, 619.
[32] S. Fleischer, Y. Khodorkovsky, E. Gershnabel, Y. Prior, I. S. Averbukh, *Isr. J. Chem.* 2012, 52, 414.
[33] E. Prost, H. Zhang, E. Hertz, F. Billard, B. Lavorel, P. Béjot, J. Zyss, I. S. Averbukh, E. Faucher, *Phys. Rev. A* 2017, 96, 043418.
[34] M. D. Poulsen, E. Peronne, H. Stapelfeldt, C. Z. Bisgaard, S. S. Viftrup, E. Hamilton, T. Seideman, *J. Chem. Phys.* 2004, 121, 783.
[35] G. Karras, M. Ndong, E. Hertz, D. Sugny, F. Billard, B. Lavorel, O. Faucher, *Phys. Rev. Lett.* 2015, 114, 103001.
[36] S. Fleischer, Y. Khodorkovsky, Y. Prior, I. S. Averbukh, *New J. Phys.* 2009, 11, 105039.
[37] K. Kitano, H. Hasegawa, Y. Ohshima, *Phys. Rev. Lett.* 2009, 103, 223002.
[38] S. Zhdanovich, A. A. Milner, C. Bloomquist, J. Floß, I. S. Averbukh, J. W. Hepburn, V. Milner, *Phys. Rev. Lett.* 2011, 107, 243004.
[39] Y. Khodorkovsky, K. Kitano, H. Hasegawa, Y. Ohshima, I. S. Averbukh, *Phys. Rev. A* 2011, 83, 023423.
[40] E. Gershnabel, I. S. Averbukh, *Phys. Rev. Lett.* 2018, 120, 083204.
[41] L. Yuan, S. W. Teitelbaum, M. Robinson, A. S. Mullin, *Proc. Natl. Acad. Sci. USA* 2011, 108, 6872.
[42] A. Korobenko, A. A. Milner, V. Milner, *Phys. Rev. Lett.* 2014, 112, 113004.
[43] J. Karczmarek, J. Wright, P. Corkum, M. Ivanov, *Phys. Rev. Lett.* 1999, 82, 3420.
[44] O. Korech, U. Steinitz, R. J. Gordon, I. S. Averbukh, Y. Prior, *Nat. Photonics* 2013, 7, 711.
[45] O. Faucher, E. Prost, E. Hertz, F. Billard, B. Lavorel, A. A. Milner, V. A. Milner, J. Zyss, I. S. Averbukh, *Phys. Rev. A* 2016, 94, 051402(R).
[46] F. A. Cotton, *Chemical Applications of Group Theory*, 3rd ed., John Wiley & Sons Ltd., Hoboken, NJ 1990.
[47] D. Hakobyan, E. Brasselet, *Nat. Photonics* 2014, 8, 610.
[48] A. Dogariu, S. Sukhov, J. Sáenz, *Nat. Photonics* 2012, 7, 24.