PHYSICAL REVIEW A **69**, 043401 (2004)

Controlling ground-state rotational dynamics of molecules by shaped femtosecond laser pulses

M. Renard, E. Hertz,* B. Lavorel, and O. Faucher
Laboratoire de Physique de l'Université de Bourgogne, UMR CNRS 5027, Boîte Postale 47 870, 21 078 Dijon cedex, France

(Received 18 November 2003; published 8 April 2004)

We report controlled excitation of ground-state rotational wave packet by pulse-shaping technique. The experiment is conducted in nitrogen (N_2) at room temperature and atmospheric pressure. A femtosecond laser pulse produces rotational coherences in the vibronic ground state of N_2 through an impulsive Raman process. The laser pulse is tailored using a spatial light modulator producing spectral phase modulation. Periodic phase steps are applied in order to control the excitation of specific rotational Raman transitions. The outcome is the modification of the relative excitation between odd and even rotational states which allows the control of the symmetry and rephasing period of the wave packet.

DOI: 10.1103/PhysRevA.69.043401 PACS number(s): 33.80.Wz, 42.50.Ct, 34.50.Rk

I. INTRODUCTION

Investigations devoted to laser-controlled chemical reactions have stimulated considerable interest in the past decades. The basic idea consists of using laser parameters as control knobs in order to drive the outcome of photoexcitation into specific final states associated with the desired reaction products. Many control schemes relying on quantum interference effects have been developed to achieve this goal. The so-called "phase control" is based, for instance, on the coherent excitation of quantum systems by a bichromatic field inducing two phase-related pathways with a different number of photons [1,2]. The excitation can be controlled through the relative phase between the two frequencies. Apart from transition probability, autoionization rate [3], angular distribution of photoelectrons and photofragments [4], as well as ionization and dissociation branching ratio [5] have been thus manipulated. With the onset of the ultrashort laser technology, new possibilities have emerged in the field of coherent control. An approach proposed by Rabitz [6] consists in producing laser pulse shapes specifically designed to control the outcome of photoreactions. Primary works have been conducted with linearly chirped laser pulses but the potentiality of such control has been greatly expanded by the advances in terms of pulse-shaping techniques [7]. Nowadays, devices producing almost any arbitrary pulse shapes are available and a large amount of experiments using "pulse shapers" have been reported. The conjunction of such control technique with feedback loop algorithm is frequently exploited in order to determine optimum laser pulse shapes when the complexity of the underlying physics prevents any reliable predictions [8–13]. Pulse-shaping techniques [14–21] as well as other approaches [22–24] have been notably applied to exercise control of ground-state molecular dynamics. The latter features major interest related to the involvement of vibrational or rotational wave packets in a wide area of quantum dynamics. The temporal evolution of rotational wave packets (RWPs) gives, for instance, access to structural information and allows diagnostics of molecular sample [25,26]. The production of a RWP with strong laser fields leads to macroscopic molecular alignment along the laser polarization [27–29]. Long-standing questions linked to orientational dependency of specific physical processes can be therefore settled by conducting analyses with aligned molecules [30]. Coherent rotational and vibrational molecular motions, impulsively [31–34] or adiabatically [35–38] excited, induce also a modulation of the refraction index well suited for temporal phase modulation. Such molecular phase modulation has been applied for spectral broadening and ultrashort optical pulse generation [31–40]. Relevance of this technique for optimization [41] of phase matching in harmonic generation has been considered too. In all these regards, investigations which aim at controlling rotational or vibrational dynamics feature appealing applications.

We demonstrate in this paper the control of rotational dynamics in the molecular vibronic ground state of N_2. The RWP is produced by a phase-shaped femtosecond laser pulse via an impulsive stimulated Raman excitation. The detection of its temporal dynamics is based on the Raman induced polarization spectroscopy (RIPS) technique. The spectral phase modulation is imprinted to the excitation field using a spatial light modulator (SLM). We have opted for judicious periodic spectral phase steps, which takes advantage of the regular energy spacing between the Raman transition frequencies. We demonstrate selective excitation of specific rotational Raman transitions. The relative even and odd rotational states' contribution to the RWP is notably controlled and, therefore, the symmetry and revival period of the wave packet.

II. EXPERIMENTAL SETUP

An overview of the experimental setup is depicted in Fig. 1. The laser apparatus is based on a chirped pulse amplified Ti:sapphire femtosecond laser. The system operates at 20 Hz repetition rate and delivers pulses of 100 fs duration around 800 nm. The output beam is split in two parts to produce the pump and probe beams. The pump passes through a pulse shaper detailed below. The relative delay between pump and probe pulse is adjusted via a corner cube retroreflector

---

*Corresponding author.
Email address: edouard.hertz@u-bourgogne.fr

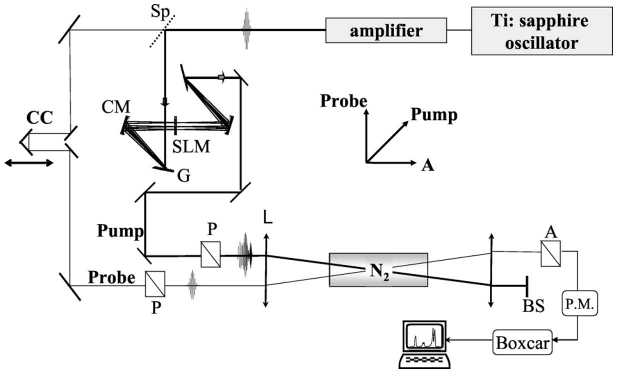

FIG. 1. Experimental setup. Sp, beam splitter; L, lens (f =175 mm); CC, corner cube; CM, cylindrical mirror (f=200 mm); G, gratings (1200 lines/mm); SLM, spatial light modulator (1×128 pixels); P, polarizer; A, analyzer; BS, beam stop; PM, photomultiplier. The relative field polarization of the pump, probe, and analyzer is depicted in the center of the figure.

mounted on a motorized linear stage. The relative polarization of the pump and probe beam is set at 45° with respect to each other via two polarizers. The two beams are then focused by a lens (f=175 mm) and cross, at small angle ($\approx 5°$), in a gas cell filled with N_2 under atmospheric pressure. Typical pump and probe energy are, respectively, 50 μJ and 2 μJ. The intensity at the laser focus for the pump and probe beam is, respectively, 4 TW/cm^2 and 0.2 TW/cm^2. At the exit of the cell, the pump beam is blocked whereas the probe passes through an analyzer (A) polarized at 90° with respect to its initial polarization. The anisotropy of the sample induced by the pump interaction is thus detected through the depolarization of the probe with a photomultiplier. The signal, sampled by a boxcar, is stored in a personal computer.

The control of the wave packet is established by tailoring the pump pulse via a conventional 4f-pulse shaper [7]. A programmable spatial light modulator is placed in the Fourier plane of the pulse shaper composed of a pair of holographic gold coated gratings and cylindrical mirrors. The spectral phase modulation of the laser can be controlled by applying appropriate voltages to the pixels. The pixel spacing is 100 μm center to center and for some reasons explained later on; the frequency spatial distribution has been finely adjusted in order to reach the desired spectral sampling of 0.79×10^{12} rad s^{-1}/pixel.

III. MODEL

As mentioned previously, the rotational wave packet is generated in the molecular vibronic ground state through an impulsive nonresonant Raman process. The laser coupling for a specific Raman transition of pulsation ω_J (with J the rotational quantum number) is proportional to

$$F(\omega_J) = \int_{-\infty}^{+\infty} E(\omega')E^*(\omega' - \omega_J)d\omega', \qquad (1)$$

denoted hereafter "Raman field excitation." $E(\omega)$ refers to the spectral electric field. By developing the latter in terms of amplitude $\varepsilon(\omega)$ and phase $\phi(\omega)$,

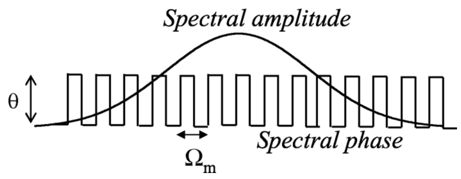

FIG. 2. Schematic representation of a periodic phase step modulation of the spectral laser phase.

$$E(\omega) = \varepsilon(\omega)\exp[i\ \phi(\omega)] \qquad (2)$$

the factor $F(\omega_J)$ becomes

$$F(\omega_J) = \int_{-\infty}^{+\infty} \varepsilon(\omega')\varepsilon(\omega' - \omega_J)\exp[i\Delta\phi(\omega')]d\omega', \qquad (3)$$

with

$$\Delta\phi(\omega') = \phi(\omega') - \phi(\omega' - \omega_J) \qquad (4)$$

the phase difference between each pair of spectral components $(\omega', \omega' - \omega_J)$ involved in the Raman excitation. According to Eq. (3), a control of both phase and amplitude of the Raman field excitation $F(\omega_J)$ is achieved by modifying the spectral laser phase. This effect, which results from quantum interference between excitation channels combining different laser frequencies, has been termed [42] "multiphoton intrapulse interference." When a periodic phase step of magnitude θ is imprinted on a symmetric spectral amplitude (Fig. 2), two specific values of the modulation period Ω_m are of a particular interest for selective excitation of Raman modes. If the Raman transition frequency ω_J is a multiple of Ω_m (i.e., $\omega_J = N\Omega_m$ with N integer), then the spectral phase difference $\Delta\phi(\omega') = 0$ independently of ω'. As a result, the excitation is similar to the one achieved with a Fourier-transform-limited pulse. Whereas if ω_J is a half-integer value of Ω_m [i.e., $\omega_J = (N+1/2)\Omega_m$], the phase difference $\Delta\phi(\omega') = \pm\theta$ and the Raman field excitation is modulated by a $\cos(\theta)$ term. These results can be summarized as follows:

$$\omega_J = N\Omega_m \quad \Rightarrow \Delta\phi(\omega') = 0, \quad \forall\ \omega' \Rightarrow F(\omega_J) = F_{TLP}(\omega_J), \qquad (5a)$$

$$\omega_J = (N + 1/2)\Omega_m \quad \Rightarrow \Delta\phi(\omega') = \pm\theta, \quad \forall\ \omega' \Rightarrow F(\omega_J) = F_{TLP}(\omega_J)\cos(\theta), \qquad (5b)$$

where $F_{TPL}(\omega_J)$ denotes the Raman field excitation at ω_J with a Fourier-transform-limited pulse, i.e., θ=0. Thus, in the conditions described by Eq. (5b), the amplitude (and sign) of Raman excitations can be controlled at any desired level by adjusting the magnitude of the phase step θ.

When an ensemble of molecules is considered, the wave packet is built from the contribution of the whole set of populated rotational states excited within the laser bandwidth. The temporal evolution of the RWP induced by the phase-shaped femtosecond laser pulse is monitored through the time-resolved polarization technique RIPS. Periodic rephasing of the RWP induces an orientational anisotropy of the sample detected by measuring the polarization rotation of

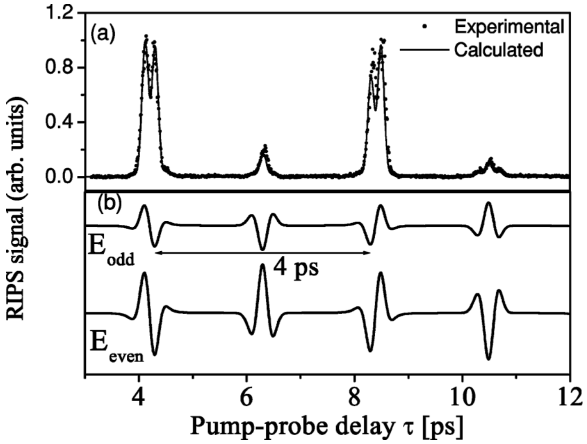

FIG. 3. (a) RIPS signal produced by an unshaped ($\theta=0$) pump pulse. (b) Contribution of odd and even rotational states to the overall signal (a). The latter is obtained by the sum squared of the odd and even contributions. The indicated arrow is used for the time-domain analysis (see text).

a time-delayed probe pulse. The signal corresponds thus to the time integral of the signal electric field squared detected through the analyzer (Fig. 1). An analytic expression of this signal as a function of the pump-probe delay τ can be written [43]:

$$I(\tau) \propto \int_{-\infty}^{+\infty} |E_{sig}(t)|^2 dt \propto \int_{-\infty}^{+\infty} \left| \Lambda(t-\tau) \mathrm{Im}\left(\sum_J T_J F(\omega_J) \times \exp(-i\omega_J t - \gamma_J t) \right) \right|^2 dt, \tag{6}$$

in which E_{sig} is the signal field. Λ refers to the temporal profile of the probe envelope, T_J is a factor depending on molecular parameters, and γ_J stands for the Raman linewidth. It should be mentioned that the factor T_J depends on the rotational quantum number J and is proportional to the nuclear spin degeneracy factor g_J.

The signal produced in N_2 with a Fourier-transform-limited pulse ($\theta=0$) is depicted in Fig. 3(a) together with the calculated trace obtained from direct computation of Eq. (6). The observed alternation of small and large transients is specific to linear molecules with a center of symmetry and a nonzero nuclear spin. It results from the difference of spin degeneracy factor between rotational states of odd- and even-J values, respectively, equal to 3 and 6 in N_2. This point is illustrated in Fig. 3(b), where the contribution to the theoretical signal of odd and even rotational states is plotted. It is shown that even rotational levels contribute to a signal field E_{even} two times larger than E_{odd}, the signal field produced by the odd distribution. As a result, the small transient recurrences observed in Fig. 3(a) arise from partial destructive interference between odd and even rotational distributions. With no alternation in the spin factor degeneracy, both odd and even rotational wave packet contributions would cancel each other out at these particular delays leading to a period of recurrence $T=1/(4Bc)=4.2$ ps (with $B=1.989$ cm^{-1} the rotational constant). It should be noted that transients observed in the signal RIPS reflect molecular alignment [28] occurring periodically subsequent to the interaction with the pump. In the present work, the extent of alignment is rather weak due to the low-field regime under which the experiment is conducted.

IV. RESULTS AND DISCUSSION

As detailed above, interference between odd and even rotational Raman modes plays a crucial role in the dynamics of the wave packet. In this respect, the controlled excitation of even rotational components, while the excitation of the odd ones remains unaffected as compared to the one produced with a Fourier-transform-limited pulse, reveals a number of attractive potentialities. This control is in principle achieved when all the even-J-value frequencies fulfilled the conditions of Eq. (5b), while the odd ones satisfy Eq. (5a). Unfortunately, such requirements cannot be strictly satisfied for all J. A satisfactory solution is to approach these criteria with the Raman transitions prevailing in the signal. With an unshaped laser pulse, the odd and even rotational levels having the prominent contribution in the RWP are $J=7$ and $J=8$, respectively. Therefore, the modulation period Ω_m should fulfill the following set of equations:

$$\omega_8 = (N+1/2)\Omega_m, \tag{7a}$$

$$\omega_7 \cong N\Omega_m. \tag{7b}$$

For a linear molecule in $^1\Sigma$ state, the pulsation of Raman transition is $\omega_J=4\pi Bc(2J+3)$ (disregarding the centrifugal distortion). For N_2 molecule, it leads to

$$\Omega_m = 16\pi Bc(19/18) = 3.16 \times 10^{12} \text{ rad/s}, \tag{8a}$$

$$N=4. \tag{8b}$$

This spectral modulation does not satisfy Eqs. (5a) and (5b) for all odd- and even-J values, respectively. Nevertheless, it provides phase difference $\Delta\phi$ close to the ones defined in these equations as far as low ($J\leqslant 7$) and high ($J \geqslant 8$) J values are not concerned. The latter two correspond indeed to intermediate cases between Eqs. (5a) and (5b). The value of the modulation period $\Omega_m=3.16\times 10^{12}$ rad/s is quite small with respect to the resolution of the 128 pixel modulator. When the spatially dispersed frequency spectrum matches the full aperture of the SLM, Ω_m covers an aperture equivalent to about 4 pixels. The frequency spatial distribution has been finely adjusted by tilting the grating angle in the shaper in order to reach the exact aperture of 4 pixels for Ω_m, corresponding to a spectral sampling of 0.79×10^{12} rad s^{-1}/pixel. We emphasize that the above procedure could be avoided by using a modulator with a larger number of pixels.

Our primary goal is to manipulate the wave packet dynamics by controlling the relative excitation between odd and even rotational distributions. As already mentioned, the occurrence of small transient recurrences in Fig. 3(a) is caused by the difference of the spin degeneracy factor for odd and even rotational levels. With the same factor, these

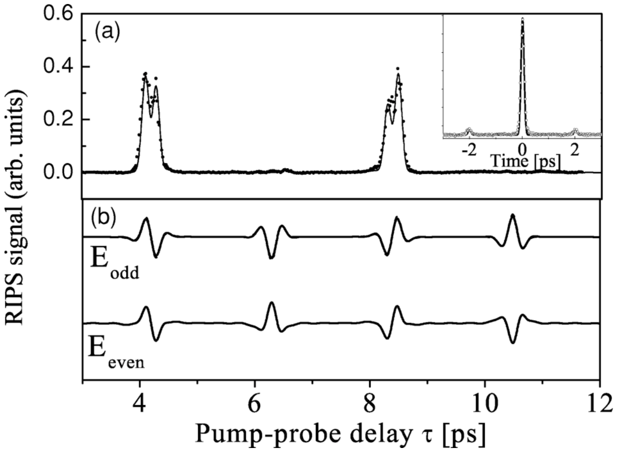

FIG. 4. (a) RIPS signal produced by a phase-shaped pump pulse $[\theta=1.18(\pi/3)]$ with the inset showing intensity cross correlation associated with this modulation. (b) Even rotational state contribution (E_{even}) is reduced by a factor of 2 compared to an exciting Fourier-transform-limited pulse (see text).

two distributions would interfere destructively at these particular delays. We propose at first to use the periodical phase modulation to balance this effect. The spin factor degeneracy for states of even-J values is twice larger than for the odd ones, and thus their excitation should be attenuated by a factor 2 with respect to the odd ones. In the conditions defined by Eq. (8), we should be able to control the Raman excitation of even-J values, while leaving almost unchanged the excitation of the odd ones. By applying a spectral modulation of magnitude $\theta=\pi/3$, the excitation of even components is in principle modified by a factor $\cos(\pi/3)=1/2$ [Eq. (5b)]. Actually, a slight deviation from this value $\theta=\pi/3$ must be considered as a result of the approached solution found in Eq. (8). The correction factor, deduced empirically by numerical simulation, leads to an amplitude modulation of $\theta=1.18(\pi/3)$. The resulting RIPS trace obtained with such spectral modulation [Fig. 4(a)] exhibits the expected transient suppression for pump-probe delays $\tau=6.3$ and 10.5 ps. The bottom panel corroborates the achievement of the sought after effect since equivalent contribution for odd and even components is observed. Perfect destructive superposition occurs for $\tau=6.3$ and 10.5 ps, leading to the modification of revival period. By modulating the spectral phase with a periodic phase step, we demonstrate therefore the possibility of controlling the rephasing properties of a rotational wave packet. The electric-field excitation synthesized by such phase modulation consists of a pulse train as shown in inset by the intensity cross correlation (displaying the intensity squared). In the same spirit, it is possible to annihilate the excitation of even rotational components by using a value $\theta\cong\pi/2$ $[\theta=1.11(\pi/2)$ precisely]. In this case, recurrences of equal amplitude are observed [Fig. 5(a)] with a half periodicity $T=1/(8Bc)=2.1$ ps. Figure 5(b) displays the sought after effect with a quasiexclusive excitation of odd rotational states. In the molecule N_2, the odd rotational levels are antisymmetric with a negative parity, whereas even are symmetric with a positive parity. The prominent excitation of odd rotational components leads thus to the production of a wave packet with a specific parity and symmetry. This aspect could be considered in investigations implying wave packets of well-defined parity and/or symmetry such as, for instance, those devoted to parity violation in molecules [44].

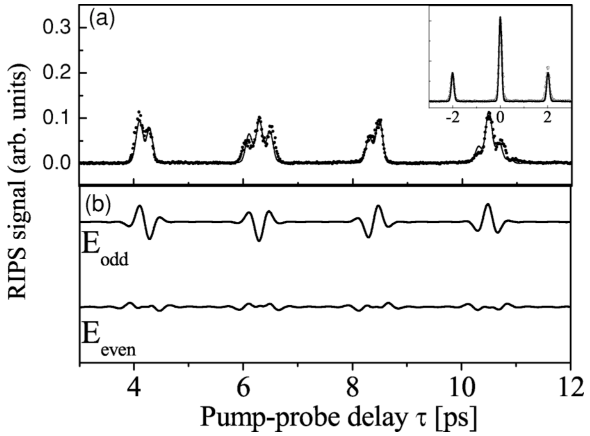

FIG. 5. (a) RIPS signal produced by a phase-shaped pump pulse $[\theta=1.11(\pi/2)]$. (b) Wave packet consists essentially of an odd rotational state contribution (see text).

The N_2 signal depicted in Fig. 3 exhibits another noticeable feature. The odd and even contributions turn out to be maximized for delays ($\tau=6.3$ and 10.5 ps) where they interfere destructively. At these particular delays, a maximum rephasing between all the even-J Raman modes on one hand, and all the odd-J Raman modes on the other hand, occurs but with a relative π-phase shift between them. It is therefore consistent to consider the possibility of rephasing these two sets of components. For this task, we have applied a modulation of magnitude $\theta=\pi$ inducing a π-phase shift between Raman transitions of opposite parities. The result, depicted in Fig. 6, displays effectively rephasing between odd and even components at proper delays. The magnitude of the signal as compared to Fig. 3(a) slightly decreases. This is caused by the nondominant Raman transitions whose frequencies do not fulfill the condition described in Eq. (5a) or (5b), as a result of the approximation carried out to deduce the final solution [Eq. (8)]. In other words, some pairs of photons

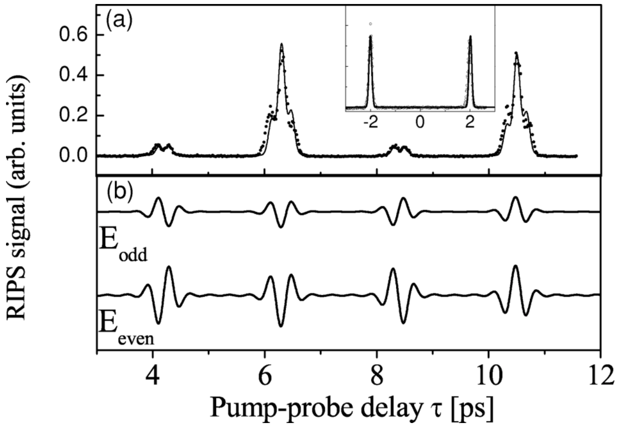

FIG. 6. (a) RIPS signal produced by a phase-shaped pump pulse ($\theta=\pi$). (b) Rephasing between odd and even rotational wave packet contribution at $\tau=6.3$ and 10.5 ps.

implied in each of these Raman transitions experience a phase difference $\Delta\phi(\omega')=0$, whereas others experience $\Delta\phi(\omega')=\pm\theta$. It should be noted, however, that the signal remains comparable with a pump pulse peak intensity reduced by a factor of about 2 [as suggested by the pulse train displayed in the cross correlation trace inserted in Fig. 6(a)]. Such spectral phase modulation features major interest for molecular alignment. Laser-induced molecular alignment is indeed intrinsically limited by the ionization process. A single pulse shaped into a train of pulses with lower energy but producing comparable alignment could be exploited in order to shift this expected limitation. This possibility will be investigated in a near future.

The frequency-domain interpretation can be supported by an analysis in the time domain [45,46]. Cross correlations inserted in Figs. 4–6 reveal that phase modulations considered in this work lead to trains of pulses, temporally shifted by $2\pi/\Omega_m$, whose relative amplitudes depend on the phase step magnitude θ. Each pulse is Fourier-transform limited and results therefore in signal fields E_{odd} and E_{even}, similar to those depicted in Fig. 3(b) $(\theta=0)$, time delayed by $2\pi/\Omega_m$ with respect to each other. For instance, modulation with $\theta=\pi$ (Fig. 6) generates mainly two identical pulses, respectively, centered at $\tau=-2$ ps and $\tau=+2$ ps. The signal field E_{odd}, for the overall sequence represented in Fig. 6(b), corresponds hence to the superposition of E_{odd} $(\theta=0,\ \tau=-2\ \mathrm{ps})$ and E_{odd} $(\theta=0,\tau=+2\ \mathrm{ps})$ while E_{even} corresponds to the superposition of E_{even} $(\theta=0,\tau=-2\ \mathrm{ps})$ and E_{even} $(\theta=0,\tau=+2\ \mathrm{ps})$ [see the arrow indicated in Fig. 3(b)]. Similar analysis can be considered for the results presented in Figs. 4 and 5.

V. CONCLUSION

In this paper, the control of rotational dynamics by pulse-shaping technique is demonstrated. The wave packet is prepared by impulsive Raman excitation in the vibronic ground state of N_2 molecule. The excitation pulse is tailored by modulating the spectral phase with a spatial light modulator. In contrast to investigations using feedback loop algorithm, we have opted for well-defined periodic phase function. The resulting manipulation of the wave packet as well as its dynamics is interpreted by means of a frequency-domain analysis. It is shown that precise control of the wave packet is possible even when using a 128 pixel modulator. The approach chosen to determine the appropriate modulation frequency of the periodic phase function is described in detail. The relative excitation between odd and even rotational components of the wave packet is controlled and thus its symmetry and rephasing properties. The production of a wave packet with a predominant antisymmetric contribution is reported. The relevance of the technique in the context of violation parity observation is mentioned. A spectral modulation which produces a rephasing of the wave packet in conjunction with a reduction of the laser pulse intensity is also investigated. It offers a valuable method for the optimization of dynamic alignment under strong field.

Manipulation of rotational coherences features also promising applications in the frame of optical diagnostics. Femtosecond time-resolved spectroscopy techniques such as CARS or RIPS have been used [25,26] to determine temperature or concentration of molecules in gas phase. Thermometry measurements rely, for instance, on the fact that transient shapes are inherently temperature dependent. However, sensitivity of transient shapes is often effective only in a delimited range of temperature and tends to decrease at high temperature [47]. By specifically tailoring the pump pulse, it is possible to induce transient revivals with an enhanced sensitivity with respect to the temperature improving thus the accuracy of measurement. This should be considered as a continuation of the present work.

ACKNOWLEDGMENTS

The authors acknowledge financial support by the Conseil Régional de Bourgogne, by the CNRS, and by the ACI “photonique” from the French Ministry of Research. Stimulating discussions with R. Marquardt are acknowledged.

---

[1] M. Shapiro, J. W. Hepburn, and P. Brumer, Chem. Phys. Lett. **149**, 451 (1988).
[2] C. Chen, Y. Yin, and D. S. Elliot, Phys. Rev. Lett. **64**, 507 (1990).
[3] D. Xenakis *et al.*, J. Phys. B **32**, 341 (1999).
[4] B. Sheehy, B. Walker, and L. F. DiMauro, Phys. Rev. Lett. **74**, 4799 (1995).
[5] L. Zhu *et al.*, Science **270**, 77 (1995).
[6] A. P. Peirce, M. A. Dahleh, and H. Rabitz, Phys. Rev. A **37**, 4950 (1988).
[7] A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).
[8] R. S. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992).
[9] C. J. Bardeen *et al.*, Chem. Phys. Lett. **280**, 151 (1997).
[10] A. Assion *et al.*, Science **282**, 919 (1998).
[11] T. Brixner *et al.*, Nature (London) **414**, 57 (2001).
[12] R. J. Levis, G. M. Menkir, and H. Rabitz, Science **292**, 709 (2001).
[13] R. Bartels *et al.*, Nature (London) **406**, 164 (2000).
[14] T. C. Weinacht, J. L. White, and P. H. Bucksbaum, J. Phys. Chem. A **103**, 10 166 (1999).
[15] T. C. Weinacht *et al.*, Chem. Phys. Lett. **344**, 333 (2001).
[16] D. Zeidler *et al.*, J. Chem. Phys. **116**, 5231 (2002).
[17] N. Dudovich, B. Oron, and Y. Silberberg, Nature (London) **418**, 512 (2002).
[18] A. M. Weiner *et al.*, Science **247**, 1317 (1999).
[19] J. G. Underwood *et al.*, Phys. Rev. Lett. **90**, 223001 (2003).
[20] D. M. Villeneuve *et al.*, Phys. Rev. Lett. **85**, 542 (2000).
[21] J. Karczmarek *et al.*, Phys. Rev. Lett. **82**, 3420 (1999).
[22] D. J. Tannor and S. A. Rice, J. Chem. Phys. **83**, 5013 (1985).
[23] I. Pinkas, G. Knopp, and Y. Prior, J. Chem. Phys. **115**, 236 (2001).
[24] J. Faeder *et al.*, J. Chem. Phys. **115**, 8440 (2001).

[25] T. Lang *et al.*, J. Chem. Phys. **115**, 5418 (2001).
[26] E. Hertz *et al.*, J. Chem. Phys. **113**, 6629 (2000).
[27] P. F. Rosca and M. J. J. Vrakking, Phys. Rev. Lett. **87**, 153902 (2001).
[28] V. Renard *et al.*, Phys. Rev. Lett. **90**, 153601 (2003).
[29] H. Stapelfeldt and T. Seideman, Rev. Mod. Phys. **75**, 543 (2003).
[30] I. V. Litvinyuk *et al.*, Phys. Rev. Lett. **90**, 233003 (2003).
[31] M. Wittmann, A. Nazarkin, and G. Korn, Phys. Rev. Lett. **84**, 5508 (2000).
[32] M. Wittmann, A. Nazarkin, and G. Korn, Opt. Lett. **26**, 298 (2001).
[33] N. Zhavoronkov and G. Korn, Phys. Rev. Lett. **88**, 203901 (2002).
[34] R. A. Bartels *et al.*, Phys. Rev. Lett. **88**, 013903 (2002).
[35] A. V. Sokolov *et al.*, Phys. Rev. Lett. **85**, 562 (2000).
[36] A. V. Sokolov, D. D. Yavuz, and S. E. Harris, Opt. Lett. **24**, 557 (1999).
[37] A. V. Sokolov, Opt. Lett. **24**, 1248 (1999).
[38] A. V. Sokolov *et al.*, Phys. Rev. Lett. **87**, 033402 (2001).
[39] V. Kalosha *et al.*, Phys. Rev. Lett. **88**, 103901 (2002).
[40] M. Spanner and M. Y. Ivanov, Opt. Lett. **28**, 576 (2003).
[41] R. A. Bartels *et al.*, Opt. Lett. **28**, 346 (2003).
[42] K. A. Walowicz *et al.*, J. Phys. Chem. A **106**, 9369 (2002).
[43] M. Morgen *et al.*, J. Chem. Phys. **102**, 8780 (1995).
[44] M. Quack, Angew. Chem., Int. Ed. **41**, 4619 (2002).
[45] E. Hertz *et al.*, Phys. Rev. A **61**, 033816 (2000).
[46] E. Hertz *et al.*, J. Chem. Phys. **113**, 6132 (2000).
[47] H. Tran *et al.*, J. Raman Spectrosc. **34**, 994 (2003).