Structural deformation of nitro group of nitromethane molecule in liquid phase in an intense femtosecond laser field*

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The structural deformation of NO_2 group induced by an intense femtosecond laser field of liquid nitromethane (NM) molecule is detected by time- and frequency-resolved coherent anti-Stokes Raman spectroscopy (CARS) technique with the intense pump laser. Here, we present the mechanism of molecular alignment and deformation. The CARS spectra and its FFT spectra of liquid NM show that the NO_2 torsional mode couples with the CN symmetric stretching mode and that the NO_2 group undergoes ultrafast structural deformation with a relaxation time of 195 fs. The frequency of the NO_2 torsional mode in liquid NM (50.8±0.3 cm^{-1}) at room temperature is found. Our results prove the structural deformation of two groups in liquid NM molecule occur simultaneously in the intense laser field.

Keywords: coherent anti-Stokes Raman spectroscopy (CARS) spectra, structural deformation, intense laser field, liquid nitromethane

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1. Introduction

The photodynamics of molecules under the influence of femtosecond, strong, and nonresonance laser pulses is now becoming a topic of wide interest.[1,2] Pulses with peak intensities of $\sim 10^{13}$ W/cm^2 generate an electric field of 1 V/Å, causing significant changes in the electronic energy level of the molecule.[3] The corresponding physical phenomena, including high-order harmonic generation, above threshold ionization, double ionization, Coulomb explosion (CE), and dissociative double ionization (DDI), have been investigated.[4] Gas NM under femtosecond laser pulses with intensities of $\sim 10^{13}$ W/cm^2 and $\sim 10^{14}$ W/cm^2 was investigated by time of flight mass spectrometer (TOFMS). NM is liquid at room temperature and atmospheric pressure. The reaction dynamics of liquid phase NM under different conditions is important for the use of NM as a detonating homogeneous liquid explosive. However, most of these experiments mentioned above have focused on gas phase NM but provided insufficient information on liquid NM. Few studies of NM in liquid phase induced by intense nonresonant femtosecond laser field have been done. This is mainly due to the restriction of experimental methods. Mass spectrometry and ion imaging have been used to determine the dissociation pathway by detecting the major, submajor, and minor products' fragment ions produced after CE. These methods require the sample to be in a gas phase.

Research from our group has been taking advantage of the coherent anti-Stokes Raman spectroscopy (CARS) technique to provide a nondestructive and nonintrusive method to detect the ultrafast dynamic process induced by nonresonance laser field in real time. The CARS technique is modified by using an intense nonresonant femtosecond pump laser (10^{11}–10^{12} W/cm^2), which has been used to drive the liquid methyl iodide (CH_3I) and NM molecules and simultaneously track their relaxation process.[5,6] In previous research, the structural deformation of CH_3 group has been detected.[6] However, the structural deformation of the NO_2 group of NM has not been detected. In fact, the structural deformation of the NO_2 group should also occur in an intense laser field. Our study has attempted to probe the evolution of ultrafast dynamic processes of the NO_2 group in liquid phase NM induced by intense femtosecond laser.

In this work, we performed the intense pumping and time- and frequency-resolved CARS on liquid NM and successfully tracked the structural deformation of the NO_2 group induced by intense nonresonant femtosecond laser pulse in real time. Additionally, we measured the value of the torsional mode of the NO_2 group in liquid NM from the process of molecular deformation. We found that the structural deformation of the two groups in the NM molecule occurred simultaneously in the intense laser field.

2. Theoretical description

Molecules orientate randomly in the liquid phase. The polarization of the electric field of laser can create an induced

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dipole moment when the molecule is in the intense laser field. A nonresonant laser field applies forces and torques to align molecules.[7] A linear molecule with a simple structure is taken as an example. When the laser interacts with the matter, the electric field component of the laser field plays a main role. To simplify the problem, the laser field is equalized in an alternating electric field along the x-axis. As shown in Fig. 1, the ellipsoid represents the linear molecule, and $\boldsymbol{E}$ is the alternating electric field of nonresonant laser field along the x-axis. The direction of $\boldsymbol{E}$ changes rapidly, and the nucleus cannot keep up with the change of $\boldsymbol{E}$. As the direction of $\boldsymbol{E}$ changes, the average effect of the intrinsic dipole moment $\boldsymbol{P}$ and $\boldsymbol{E}$ is offset. The quality of electrons is very small, so the direction of the $\boldsymbol{E}$ has an effect on the distribution of electron cloud. This will change the relative position of the molecular skeleton, and the molecule will produce the induced dipole moment $\Delta\boldsymbol{P}$ which is proportional to $\boldsymbol{E}$

$$\Delta\boldsymbol{P} = \alpha\boldsymbol{E}, \tag{1}$$

where α is the polarizability. The torque $\boldsymbol{M} = \Delta\boldsymbol{P} \times \boldsymbol{E}$ aligns the molecule along the direction of $\boldsymbol{E}$.

When the laser interacts with the molecule, the Hamiltonian of the molecule can be written as

$$H = H_{\text{mol}} + H_{\text{ind}}, \tag{2}$$

where H_{mol} is the Hamiltonian of the molecule without an external field, and H_{ind} is the interaction of the laser and the induced dipole moment

$$H_{\text{ind}} = -\frac{1}{4}\sum_{\rho,\rho'} \varepsilon_\rho \alpha_{\rho\rho'} \varepsilon_{\rho'}, \tag{3}$$

where ρ and ρ' are space fixed Cartesian coordinate system.

The laser field can be expressed as

$$\varepsilon = \varepsilon(t)\cos(\omega t), \tag{4}$$

where $\varepsilon(t)$ is the envelope of electric field, and ω is angular frequency of laser. For the linear molecule

$$\alpha_{XX} = \alpha_{\parallel}, \tag{5}$$

where $\alpha_{\parallel}$ is the projection of polarizing onto the molecular axis.

$$\alpha_{ZZ} = \alpha_{YY} = \alpha_{\perp}, \tag{6}$$

where $\alpha_{\perp}$ is the projection of polarization onto the molecular axis.

When the laser field is polarized

$$\varepsilon = \varepsilon_x, \tag{7}$$

the interaction of the linear molecule and laser field depends on the angle between $\Delta\boldsymbol{P}$ and $\boldsymbol{E}$. Equation (3) can be simplified into

$$H_{\text{ind}} = -\frac{1}{4}\varepsilon^2(t)(\alpha_{\parallel} - \alpha_{\perp})\cos^2(\theta), \tag{8}$$

where θ is the angle between $\Delta\boldsymbol{P}$ and $\boldsymbol{E}$. Under the effect of the electric field component of the laser, the molecule tends to move to the position with the lowest energy. According to Eq. (8), the position with the lowest energy corresponds to $\theta = 0$. This means that molecular axis will be the same as the direction of electric field component of the laser.

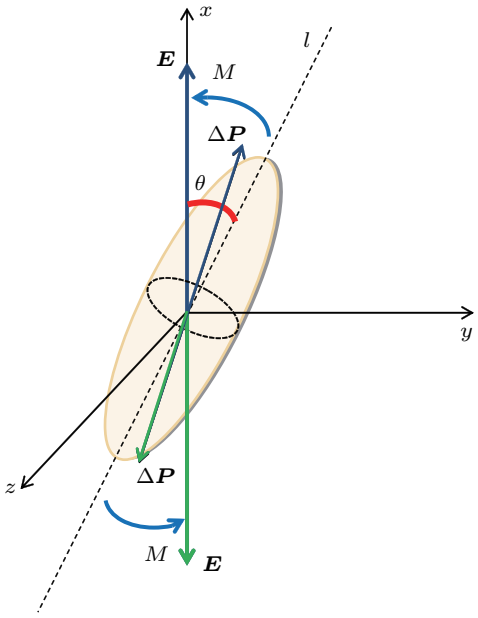


Fig. 1. (color online) The alignment of linear molecule induced by the intense laser field.

The intense nonresonant laser field can also induce the molecular structural deformation. As shown in Fig. 2, the NO_2 group of the NM molecule is taken to show the principle of structural deformation induced by the intense nonresonant laser field. The torque $M = \Delta\boldsymbol{P} \times \boldsymbol{E}$ can align the N–O bond to the direction of $\boldsymbol{E}$. Therefore, the O–N–O angle decreases within the pulse duration as shown in Fig. 2. After the pump pulse passes by, the structure relaxes gradually to the equilibrium conformation.

The mechanism of molecular deformation induced by the nonresonant intense laser field can also be explained by the quantum mechanism. In a typical Raman process, the interaction of pump pulse and Stokes pulse at the zero time leads to an additional interaction. This can be expressed as[8]

$$\begin{aligned} V = &-\frac{1}{4}(\varepsilon_{\text{p}}^* \cdot \boldsymbol{\alpha} \cdot \varepsilon_{\text{p}} + \varepsilon_{\text{S}}^* \cdot \boldsymbol{\alpha} \cdot \varepsilon_{\text{S}} + \varepsilon_{\text{p}}^* \cdot \boldsymbol{\alpha} \cdot \varepsilon_{\text{S}} \mathrm{e}^{-\mathrm{i}\omega_{\text{pS}}t} \\ &+ \varepsilon_{\text{S}}^* \cdot \boldsymbol{\alpha} \cdot \varepsilon_{\text{p}} \mathrm{e}^{\mathrm{i}\omega_{\text{pS}}t}), \end{aligned} \tag{9}$$

where ε_{p} is the envelope of pump pulse, ε_{S} is the envelope of Stokes pulse, and ω_{ps} is the difference frequency. Equation (9)

can be written as

$$V = V_{\rm p} + V_{\rm S} + V_{\rm pS} + V_{\rm Sp}, \tag{10}$$

where $V_{\rm p}$ is the additional interaction of the intense pump laser, $V_{\rm S}$ is the additional interaction of Stokes laser, and $V_{\rm pS}$ and $V_{\rm Sp}$ are the interaction of coherent vibrational energy lever.

In our experiment, the intensity of pump pulse is much greater than that of Stokes pulse, so $V_{\rm S}$, $V_{\rm pS}$, and $V_{\rm Sp}$ can be ignored. The effect of $V_{\rm p}$ equates to making the molecular potential energy surface dressed. The "dressed energy potential" means that the molecular potential energy surface deforms, and the atoms can move quickly on the "dressed energy potential". This leads to molecular deformation.

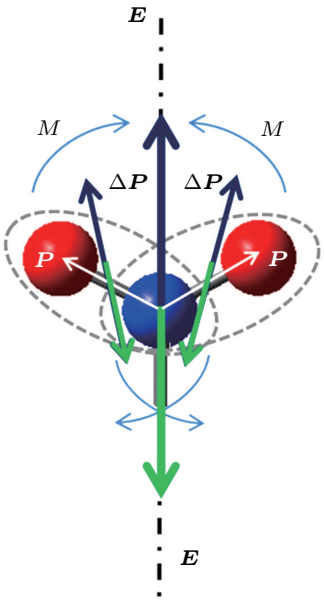


Fig. 2. (color online) Schematic of the mechanism for molecular deformation of NO_2 group. The blue and red balls denote nitrogen and oxygen atoms, respectively.

3. Experiment

The overall experimental setup used for the femtosecond CARS is given in Fig. 3. A 110 fs, 1.0 mJ, and 800 nm pulse from a 1 kHz Ti: sapphire amplifier was focused on the sample of liquid NM filling a quartz glass cuvette of 1 mm thickness. The laser pulse is divided into three parts by two beam splitters (BS). Two parts were used as the pump and probe pulses. The third one produced a supercontinuum (SC) pulse as the Stokes pulse by passing an Al_2O_3 crystal.[9] The SC pulse had an ultrabroadband spectral profile, ranging from 400 nm to 1100 nm. The pump pulse was controlled by an optical attenuator to adjust the pump pulse energy. The intensity of pump energy was chosen to be 0.33, 0.66, 1, 2, 3, and 5 μJ. The pump pulse was focused by a 175 mm focal length lens. By assuming a Gaussian spatial profile, the spot size of the laser beam was estimated to be 70 μm in diameter. Therefore, the power densities are 7.9×10^{10}, 1.6×10^{11}, 2.4×10^{11}, 4.8×10^{11}, 7.2×10^{11}, and 1.2×10^{12} W/cm^2, respectively. The probe pulse was chosen to be femtosecond pulse. The intensities of Stokes and probe were set much lower than that of the pump pulse. Two delay lines were motorized by a motion controller. One delay line was used to produce the temporal overlap of the pump and Stokes pulse by changing the delay time between the pump and SC pulse. Another one provided the variable time delay for the probe pulse. All three beams were sent through a lens focused onto the sample, and a collecting lens sent the signal to the CCD spectrometer. The folded BOXCARS geometry was used, in which angles, properly chosen between the beams, were determined by the four-wave-mixing phase-matching condition. The signal generated in a direction $\boldsymbol{k}_{\rm CARS} = \boldsymbol{k}_{\rm pu} - \boldsymbol{k}_{\rm St} + \boldsymbol{k}_{\rm pr}$.[10] The CN symmetric stretching mode, which is the characteristic vibrational mode, of the NM molecule (917 cm^{-1}), was chosen to be excited. All measurements were taken at room temperature.

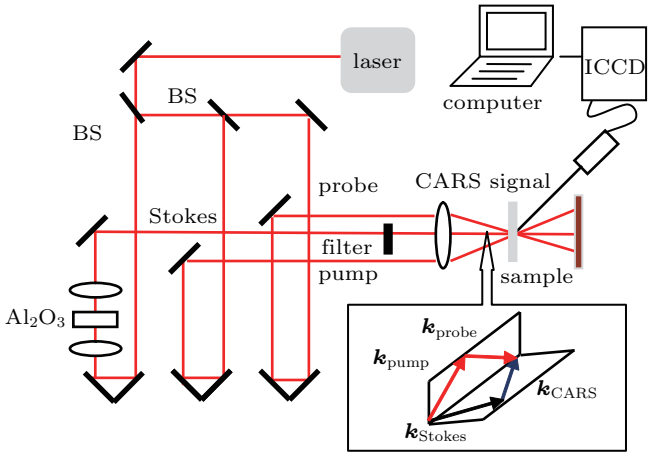


Fig. 3. (color online) Schematic diagram of the experimental setup for the intense pumping and time- and frequency-resolved CARS. BOXCARS configuration was used in the experiment.

4. Results and discussion

The analytical pure NM was used in the experiment. The CARS signal was centered at approximately 917 cm^{-1}(the CN symmetric stretching mode) and covered the region from 700 to 1050 cm^{-1}. Figure 4 shows the pump pulse intensity-dependent frequency resolved CARS spectra at a selected delay time of 1200 fs. The pulse energies were 0.33, 0.66, 1, 2, 3, and 5 μJ. When the pump pulse energy was in the low-intensity range (<3 μJ), the spectral line profile was a broad envelope. These results are consistent with those of conventional CARS experiment on NM reported in the literature.[11] However, the fine structures of spectra appear when the pump pulse energy was increased to ~ 3 μJ. This is a typical feature of stimulated scattering. The signal intensity also increased with the enhancement of the pump pulse energy, which is shown in Fig. 5. If the pulse energies of the Stokes and probe pulses are fixed, the signal intensity should have a linear relationship with the pump pulse energy when only the pump

pulse energy is changed. However, the experimental results show a nonlinear relationship between the signal intensity and the pump pulse energy. When the pump pulse energy was more than 3 μJ, the signal intensity exhibited a dramatic increase. This is also a typical feature of the simulated scattering process. At the zero delay time, the pump pulse and the broadband Stokes pulse of SC prepare coherent vibrational modes. When the pump pulse energy was intense enough, the density of the coherent vibrational mode in the vibrational excited state can be much higher than that in the ground state. The intense pump pulse produces much higher density Raman mode, which leads to stimulated Raman scattering. Thus, after the zero delay time, the scattering process for the probe laser pulse should exhibit a stimulated Raman process in essence. This is proved by the pump pulse intensity-dependent experimental results.

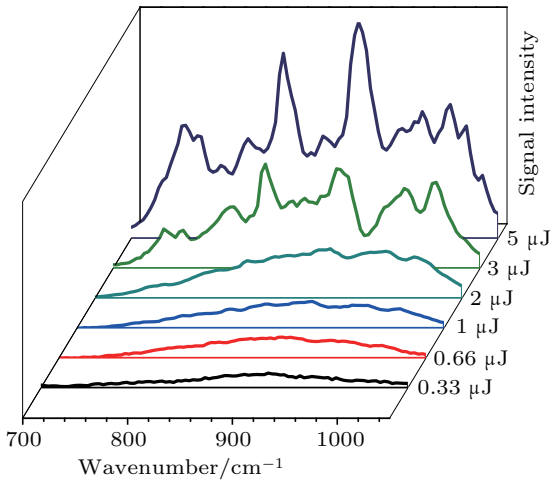


Fig. 4. (color online) Spectra obtained at different pump energies (0.33, 0.66, 1, 2, 3, and 5 μJ) at the delay time of 1200 fs. Fine structures of spectra appear when the pump pulse energy is increased to 3 and 5 μJ. The signal intensity increases with the enhancement of the pump pulse energy.

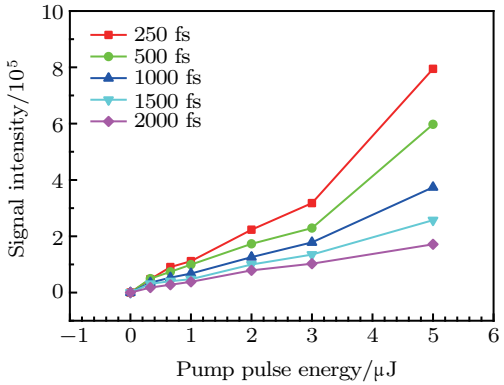


Fig. 5. (color online) Relationship between the pump pulse energy and the signal intensity at the delay times of 250, 500, 1000, 1500, and 2000 fs. The signal intensity of the spectra shows a dramatic increase when the pump pulse energy is increased to 3 and 5 μJ.

The contour plot of time- and frequency-resolved CARS spectra of liquid NM of pump intensity of 5 μJ is depicted in Fig. 6. The CARS signal intensity is formulated as[12]

$$S_{\mathrm{c}}(\tau) \propto \int_{-\infty}^{+\infty} \left| P_{\mathrm{c}}^{(3)}(\tau, t) \right|^2 \mathrm{d}t, \tag{11}$$

where $P_{\mathrm{c}}^{(3)}$ is the nonlinear polarization, and τ is the delay time between the probe pulse and the synchronous pump and Stokes pulses. The intense signal at zero time delay is the electronic nonresonant part. After approximately 100 fs, the CARS spectra had a negligible nonresonant contribution. The signal after 100 fs showed a complex temporal behavior and decayed within approximately 4 ps. After 1100 fs, the contour plot clearly showed the dephasing of the six Raman modes centered at 1006, 966, 917, 866, 800, and 750 cm^{-1}. All these Raman modes clearly underwent a frequency shift and gradually became relatively stable. According to the spontaneous Raman modes of NM, the Raman mode centered at 917 cm^{-1} was assigned to be the CN symmetric stretching (ν_{s}(CN)) mode.[13] It displayed the most intense Raman resonance and the longest dephasing time, which is consistent with the results of CARS experiment on NM reported in Ref. [11]. The other Raman modes were distributed evenly at the two sides of the position of 917 cm^{-1}. The interval of the adjacent Raman modes kept decreasing with the time delay, and stabilized after the delay time of approximately 1100 fs. Then, the average energy interval between adjacent Raman modes was approximately 51 cm^{-1}.

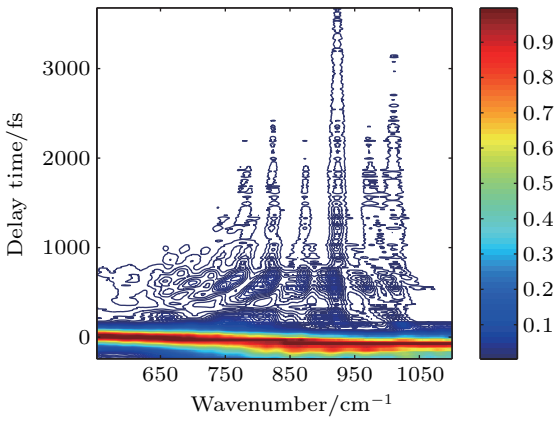


Fig. 6. (color online) Contour plot of time- and frequency-resolved CARS spectra of liquid NM when the pump intensity is 5 μJ. The two axes represent delay time and Raman vibrational frequency values, while contours represent the normalized intensity of the CARS signal. The peak intensity of every spectral position has been normalized to 1.

The torsional vibration is the restricted rotation of a part of a molecule with respect to the molecular skeleton. The frequency of the torsional mode of the NO_2 group can be denoted by ν_{Tors} (NO_2). It describes the restricted rotation of the NO_2 group of NM about the molecular axis of the C–N bond. The intense ultrashort laser pulses can induce torsional motion in a molecule.[14] At the same time, the CN symmetric stretching mode (917 cm^{-1}) is excited coherently by pump

and Stokes pulse, and the torsion–vibration coupling occurred. In Fig. 4, the converging peaks on both sides of 917 cm^{-1} were assigned as the sum and difference combinations of the torsional mode and the CN symmetric stretching mode, i.e., $\nu_s(CN) \pm n\nu_{Tors}(NO_2)$, where n $(= 0,1,2)$ was the quantum number of torsional vibration. The average energy interval between adjacent Raman modes represents the frequency value of the torsional mode. After the delay time of 1.1 ps, the value of $\nu_{Tors}(NO_2)$ was approximately 51 cm^{-1} and kept stable.

The value of $\nu_{Tors}(NO_2)$ decreased with the delay time when the structure of the NM molecule was deforming. At the zero delay time, the ultrafast structural deformation of the NO_2 group from the equilibrium conformation showed a decreased O–N–O angle induced by intense femtosecond pump pulse. The change of the O–N–O angle led to the change of rotational inertia along C–N bond. The deformed shape of the NO_2 group at the zero delay time gave the minimum rotational inertia of the C–N bond. In addition, the frequency of the NO_2 torsional mode reached a corresponding maximum. After the intense femtosecond pump pulse passed through the sample, the shape of the NO_2 group relaxed to the equilibrium state and was gradually accompanied by an increase in the rotational inertia. This caused the frequency of the NO_2 torsional mode to decrease with the delay time. This is why the intervals of the adjacent Raman modes decrease with the delay time starting from zero. After 1.1 ps, the interval of the adjacent Raman modes reached a stable state, indicating that the molecule structure recovered to the equilibrium state.

Further confirmation of the structural deformation of the NM molecules was made by fast Fourier transformation (FFT) spectra. Transforming Eq. (12) results in

$$H(\omega, \omega_{FFT}) = \hat{F}\{S_c(\omega, t)\}, \tag{12}$$

where ω gives the frequency of Raman vibrational mode at which the transient is considered, and ω_{FFT} gives the beat frequency between two adjacent Raman vibrational modes. Considering two adjacent Raman vibrational modes j and k, the beat between them can be described as

$$H(\omega_j, |\omega_j - \omega_k|) = C\exp[i(\phi_j - \phi_k)], \tag{13}$$

where $(\phi_j - \phi_k)$ is the phase shift of the beat between j and k modes, ω_j and ω_k are frequency values of j and k, and $\omega_{FFT} = |\omega_j - \omega_k|$ is the beat frequency.[15] If the FFT of the time domain signal is performed at every Raman spectral position, we will obtain two-dimensional FFT spectra of the coherent coupling of all the Raman vibrational modes. The peak of the spectral curve in the slice along the beat frequency axis in FFT spectra represents the coherence between two adjacent vibrational modes, and the peak value on the beat frequency axis is equal to their frequency difference. If the molecular structure does not change with time delay, the frequency difference ω_j–ω_k should be constant. Therefore, the spectral band due to the coupling of modes j and k in FFT spectra is parallel to Raman vibrational frequency axis. In traditional FFT spectra of time- and frequency-resolved spectra, the band features mentioned above are often observed.[16] However, if the molecular structure is unstable, ω_j and ω_k are not constants, the frequency difference ω_j–ω_k changes with the delay time, and the bands in the FFT spectra are not parallel to the Raman vibrational frequency axis. The FFT spectra of our CARS experiment are shown in Fig. 7. In data processing, we only performed a subtraction of the exponentially decaying component of the transient signal, which retained only oscillatory components. There was no additional processing or smoothing of the data. Since the nonresonant background was not of interest to our investigation, it was ignored in the data analysis by only plotting the FFT plot obtained for delay times longer than 100 fs. Some sloping bands, which were not parallel to the Raman vibrational frequency axis, appear in Fig. 7. According to the previous analysis of FFT spectra, these sloping bands indicate that the frequency of one or both adjacent vibrational modes participating in coherences is changing. This result means that the molecule structure is deformed.

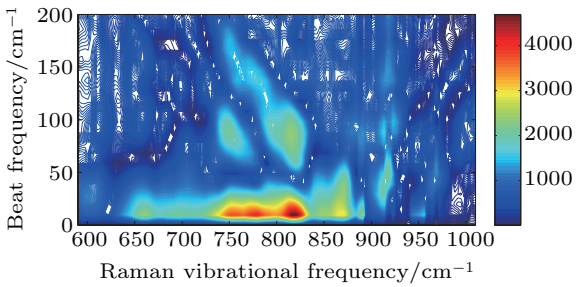


Fig. 7. (color online) Two-dimension FFT spectra. The two axes represent the beat frequency and the Raman vibrational frequency values.

The restoration time of the structural deformation of the NO_2 group can be obtained by the relaxation of the frequency of the torsional mode. The following empirical exponential function is used:

$$\nu_{Tors}(\tau) = \nu_{Tors} + \Delta\nu_{Tors}\exp(-k\tau), \tag{14}$$

where ν_{Tors} is the frequency of the torsional mode at stable molecular structure, $\Delta\nu_{Tors}$ is the torsional mode frequency variation, k is the exponential rate of the torsional mode frequency changing, and τ is the delay time. The data was analyzed from the delay time $t = 400$ fs to avoid the intense nonresonant contribution at time zero. The decay curve of the torsional mode is presented in Fig. 8, and the corresponding fitted results are $\nu_{Tors} = 50.8 \pm 0.3$ cm^{-1}, $\Delta\nu_{Tors} = (36 \pm 3) \times 10$ cm^{-1}, and $k = (51 \pm 2) \times 10^{-4}$ fs^{-1}. The fitted results indicate that the frequency of the torsional mode decays to $\sim 50.8 \pm 0.3$ cm^{-1} at the stable molecular structure which is consistent with the experimental value of the single crystals of

solid NM by means of Raman scattering (52 cm^{-1}).[17] Commonly, the exact value of the torsional mode of liquid NM is unknown. This is because at room temperature, the residence time becomes lower than the characteristic time linked to the phonon bands, and thus the torsional transitions are not observed in Raman experiments at room temperature. In our experiment, the torsional mode of liquid NM can be calculated. The timescale of the molecular structural restoration process is $\sim$ 195 fs, as obtained from k.

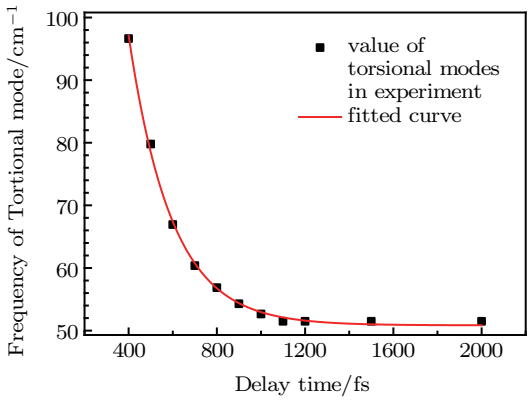


Fig. 8. (color online) Decay of the frequency of the torsional mode in experiment (black scatter) and the fitted curve (red solid curve).

5. Conclusion

In summary, the intense pumping time- and frequency-resolved CARS technique with the femtosecond probe laser successfully monitors the structural deformation of the NO_2 group of liquid NM molecules induced by intense nonresonant femtosecond laser in real time. The time- and frequency-resolved CARS spectra provided information on ultrafast structural deformation of the NO_2 group, and the restoring time was approximately 195 fs. Further confirmation of the structural deformation of the NM molecules is made by two-dimensional FFT spectra. The frequency of the NO_2 torsional mode in liquid NM (50.8 ± 0.3 cm^{-1}) at room temperature was found. On the basis of previous studies,[15] our results prove that the structural deformation of the two groups in the NM-type molecule occurs simultaneously in the intense laser field. The CARS experiment with the intense pump pulse is a feasible method to investigate reactions of liquid phase molecules induced by femtosecond laser field without complex experimental systems.

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