

Measurement of field-free molecular alignment by cross-defocusing assisted polarization spectroscopy

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Abstract: We demonstrate the measurement of field-free molecular alignment of nitrogen by a cross-(de)focusing assisted polarization spectroscopy technique, allowing us to distinguish the transient orientation of prealigned molecules on the basis of the relative ratio of the adjacent revival peaks of the measured signal. For impulsive excitation with high pump intensities, the involved orientation-dependent ionization of the neutral molecules contributes to the signal.

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1. Introduction

Field-free molecular alignment [1–4] with periodic revivals was observed in response to the quantum wake of the impulsive rotational Raman excitation by an ultrashort laser pulse. There has been an ever growing interest in studying the field-free molecular alignment as well as the related applications in various fields [5–16] by using the controllable quantum wake. The degree of molecular alignment is quantified by the average term $\langle\langle\cos^2\theta\rangle\rangle$, where θ is the angle between the molecular axis and the laser field polarization. The field-free molecular alignment could be measured with the weak field polarization technique [3] or by using the spatial cross-defocusing of the probe pulse [17,18] to obtain a homodyne signal proportional to $(\langle\langle\cos^2\theta\rangle\rangle-1/3)^2$. Alternatively, pure heterodyne signal of the field-free molecular alignment directly proportional to $\langle\langle\cos^2\theta\rangle\rangle$ could be obtained through Coulomb explosion of prealigned molecules [19]. A modified weak field polarization technique was demonstrated [20] to obtain pure heterodyne molecular alignment signal proportional to $(\langle\langle\cos^2\theta\rangle\rangle-1/3)$. Recently, it was shown that [21] the degree of molecular alignment could also be measured by using an improved single-shot supercontinuum spectral interferometry.

In this paper, we demonstrate that the field-free transient orientation of nitrogen molecules can be distinguished by combining the cross-(de)focusing and polarization spectroscopy techniques. The cross-(de)focusing effect is extracted by detecting a small part of the probe beam, which alters the polarization spectroscopy measurement in a different way for various transient orientations.

2. Experimental setup

Experimentally, an output from the amplified Ti:Sapphire laser system (pulse duration T_p : 60 fs, central wavelength λ_0 : 800 nm, repetition rate: 1 kHz, beam diameter: 7 mm at $1/e^2$) was split in two parts: the transmitted pump pulse for molecular alignment and the reflected probe pulse for signal analysis. The field polarization of the probe was rotated by 45° with respect to the pump. Both the pump and probe pulses were focused by a lens of $f = 100$ cm into a 10-cm long gas cell at a noncollinear crossing angle of $\sim 2^\circ$, which was filled with pure N₂ at a gas pressure of 1 atm. The interaction distance between the pump and probe beams was estimated to be $d = 4\sim 5$ mm for a beam waist $w_0 = 70\sim 80$ μm around the focus. The f number of the beam was then calculated to be $0.5(\pi Z_0/\lambda_0)^{1/2} \sim 143$, where $Z_0 \sim 2$ cm is the Rayleigh length [12]. Only the field component with polarization perpendicular to the incident one of the probe pulse was selected by an α -BBO polarizer (extinction ratio $\sim 1:10^{-6}$), which was then diffracted freely onto a photodiode with a sensitive area of ~ 0.4 mm² (locating at a distance of $L \sim 30$ cm away from the interaction zone). Therefore, only a small part at the beam center of the probe pulse with modulated polarization was measured, which composed of our hybrid technique with respect to the conventional weak field polarization technique [3]. The output signal from the photodiode was analyzed with a Lock-in amplifier (SR830). By using a variable attenuator, the pump pulse energy could be adjusted from 0.05 to 1.2 mJ, while the probe one was fixed to be 20 μJ to avoid any undesired self-acted nonlinear effects.

3. Theoretical model

The principle of polarization spectroscopy for molecular alignment measurement [3] is based on polarization modulation of the linearly polarized probe pulse due to the orientation-

dependent refractive index of the prealigned molecules. Since the Gaussian-shaped transverse profile of the pump pulse induces a larger molecular alignment in the beam center than in its periphery, the probe pulse meanwhile experiences an additional cross-(de)focusing effect from the prealigned molecules [10–18]. The prealigned molecular gas with an effective thickness of d is equivalent to a lens of a nonlinear focal length [17]:

$$f_m(t) = n_0 w_0^2 / [4d\Delta n_0(t)], \quad (1)$$

where n_0 is the linear refractive index, $\Delta n_0(t) = (2\pi N_0 \Delta\alpha / n_0) [\langle\langle \cos^2\theta(t) \rangle\rangle_r = 0 - 1/3]$ is the refractive index change at the beam center induced by molecular alignment, N_0 is the number density of the molecular gas, and $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$ is the polarizability difference between the components parallel (α_{\parallel}) and perpendicular (α_{\perp}) to the field polarization [2]. Therefore, when the molecules are oriented parallel or perpendicular to the field polarization with $\langle\langle \cos^2\theta \rangle\rangle$ greater or smaller than 1/3, the prealigned molecules act as a positive or negative lens, respectively. For N_2 at 1 atm with a polarizability difference of $\Delta\alpha = 1.0 \text{ \AA}^3$ excited by a pump intensity of $I_p = 4.0 \times 10^{13} \text{ W/cm}^2$ with $|\langle\langle \cos^2\theta \rangle\rangle - 1/3| \sim 0.06$, the nonlinear focal length f_m and molecular number $(8\Delta n_0)^{1/2}$ are respectively estimated to be $\sim 3 \text{ cm}$ and 115, characterizing the importance of the molecular alignment induced cross-(de)focusing effect [12].

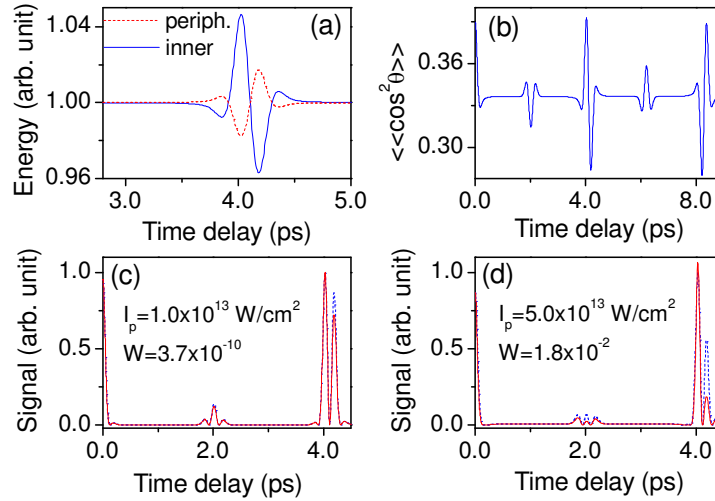


Fig. 1. (a) Modulation of the contained energy in a small aperture at the inner (solid curve) and periphery (dashed curve) of the beam profile. (b) Calculated molecular alignment metric $\langle\langle \cos^2\theta \rangle\rangle$ of N_2 versus time delay. (c), (d) Simulated output signal from the photodiode without (dashed curves) and with (solid curves) the contributions of cross-defocusing ($\eta = 5.0$) and anisotropic ionization under various pump intensities.

In order to have a more intuitive picture on the energy modulation of the probe beam induced by the additional cross-(de)focusing effect, we calculated the waist of the probe beam on the surface of the detector at a distance of L from the crossing point by assuming its propagation obeys the basic Gaussian law, which is valid for the fairly low probe pulse energy as considered here. When the pump pulse was turned off (i.e. without molecular alignment), the probe beam waist reads $w_L = w_0 [1 + L^2/Z_0^2]^{1/2}$. However, when the molecular alignment induced cross-(de)focusing effect with an equivalent focal length of f_m is included, the probe beam waist becomes to be

$$w_{Lm} = w_0 \beta [1 + [L / (Z_0 \beta) - Z_0 / f_m]^2]^{1/2}, \quad (2)$$

where $\beta = (1 + Z_0^2/f_m^2)^{-1}$. By assuming the focal length f_m of the equivalent lens induced by the molecular alignment is much greater than the Rayleigh length Z_0 (i.e. $\beta \sim 1$ and $Z_0^2/f_m^2 \sim 1$), as

compared to the case without molecular alignment of w_L , the beam waist of the probe pulse is modulated to be $w_{Lm}(t) = [w_L^2 - 2w_0^2 L f_m(t)]^{1/2}$. For a Gaussian beam with a transverse intensity distribution of $I(r, L) \sim [w_0/w_{L(m)}(L)]^2 \exp\{-2[r/w_{L(m)}(L)]^2\} \sim [w_0/w_{L(m)}(L)]^2 \{1 - 2[r/w_{L(m)}(L)]^2\}$, the energy contained at the beam center within a diameter of D can be expressed as

$$S_D(t) \sim 2 \int_0^{D/2} I(r, t) dr \sim [w_0 / w_{L(m)}]^2 \{1 - D^3 / [12w_{L(m)}]^2\}. \quad (3)$$

Therefore, when the diameter D is much smaller than the beam waist $w_{L(m)}$, the measured signal reads:

$$S_D(t) \sim 2 \int_0^{D/2} I(r, t) dr = \frac{S_{D0}}{1 - \eta[\langle\langle \cos^2 \theta(t) \rangle\rangle_{r=0} - 1/3]}, \quad (4)$$

where S_{D0} is the measured signal without molecular alignment, and $\eta \sim 4dZ_0^2 N_0 \Delta \alpha / [(Z_0^2/L + L)w_0^2]$ accounts for the cross-(de)focusing related to the molecular alignment. As shown in Fig. 1(a), such modulation of the energy contained in a small aperture along the transverse beam profile was confirmed by numerically solving the propagation equation of the probe pulse in a prealigned molecular gas [10]. It varied in accordance with the revival of the molecular alignment as shown in Fig. 1(b).

For high pump intensities, the molecular ionization during the pump excitation is unavoidable. Since a diatomic molecule is much easier to be ionized as its molecular axis is parallel to the field polarization than the perpendicular case [19,22] for its double-well potential structure, an orientation-dependent depletion of the neutral molecules is resulted as $N_k(\theta) = N_0 W_k(\theta)$, where

$$W_k(\theta) = \sigma_{K0} I_p^K T_p (a + b \langle\langle \cos^2 \theta \rangle\rangle) = W_0 (a + b \langle\langle \cos^2 \theta \rangle\rangle) \quad (5)$$

is the orientation-dependent ionization rate [6], $\sigma_{K0} = 6.31 \times 10^{-140} \text{ s}^{-1} \text{ cm}^{22} / \text{W}^{11}$ is the ionization cross-section of the randomly oriented N_2 at 800 nm, $K = 11$ is the number of photons needed to liberate an electron from N_2 , and the molecular alignment metrics are considered up to the second order.

The refractive index proportional to the neutral molecular number density $N_0[1 - W_k(\theta)]$ is thus modulated. The refractive index parallel to the field polarization of the pump pulse can be written as [23]

$$\begin{aligned} n_{//} &= 1 + 2\pi N_0 [1 - W_k(\theta)] (\alpha_{\perp} + \Delta\alpha \langle\langle \cos^2 \theta \rangle\rangle) \\ &\sim 1 + 2\pi N_0 \{ \alpha_{\perp} (1 - aW_0) + [\Delta\alpha(1 - aW_0) - bW_0 \alpha_{\perp}] \langle\langle \cos^2 \theta \rangle\rangle \}. \end{aligned} \quad (6)$$

Similarly, the modulated refractive index perpendicular to the pump polarization can be written as

$$\begin{aligned} n_{\perp} &= 1 + 2\pi N_0 [1 - W_k(\theta)] (\alpha_{\perp} + \Delta\alpha \langle\langle \sin^2 \theta \sin^2 \phi \rangle\rangle) \\ &\sim 1 + \pi N_0 \{ (\alpha_{\perp} + \alpha_{//}) (1 - aW_0) + [\Delta\alpha(aW_0 - 1) - bW_0 (\alpha_{\perp} + \alpha_{//})] \langle\langle \cos^2 \theta \rangle\rangle \}, \end{aligned} \quad (7)$$

where the relationship of $\langle\langle \sin^2 \theta \sin^2 \phi \rangle\rangle = (1 - \langle\langle \cos^2 \theta \rangle\rangle) / 2$ was used. It therefore leads to a corrected refractive index difference as:

$$\Delta n = n_{//} - n_{\perp} \sim N_0 \Delta \alpha \pi [bW_0 / 3 + (3 - 3aW_0 + bW_0) (\langle\langle \cos^2 \theta \rangle\rangle - 1/3)]. \quad (8)$$

For the case without ionization $W_0 = 0.0$, it reduces to the conventional one and leads to molecular alignment signal proportional to $(\Delta n)^2 \sim (\langle\langle \cos^2 \theta \rangle\rangle - 1/3)^2$ [3]. Since the measured molecular alignment signal by the weak field polarization spectroscopy is directly proportional to the square of the refractive index difference $(\Delta n)^2$ [3], the anisotropic depletion of the neutral molecules induced by the orientation-dependent ionization rate of the

diatomic molecules during the pump excitation also contributes the measured molecular alignment signal.

When both the cross-(de)focusing and anisotropic molecular ionization are considered, the measured signal is therefore corrected to be

$$S_D \sim (\Delta n \pi d / \lambda_0)^2 [1 - \eta \langle \cos^2 \theta \rangle - 1/3]^{-1}. \quad (9)$$

The revival peaks are expected to relatively decrease for the perpendicular orientation revivals with $\langle \cos^2 \theta \rangle - 1/3 < 0$ as compared to the parallel orientation revivals with $\langle \cos^2 \theta \rangle - 1/3 > 0$, leading to additional modulation of the measured molecular alignment signal so as to distinguish its transient orientation. This makes our hybrid technique assisted with the additional cross-(de)focusing effect and orientation-dependent ionization rate to measure the impulsive molecular alignment of the diatomic molecules quite different from the previous ones [1–4,17–21], showing its unique features and potential applications. Here, differing from the spatial defocusing from the plasma wake [17,18], the ionization correction comes from the orientation-dependent depletion of the neutral molecules during the pump excitation which modulates the polarization spectroscopy measurement. The ionization induced plasma itself with isotropic refractive index property will not alter the polarization of the probe pulse and the measured molecular alignment signal.

Figures 1(c) and 1(d) show the calculated molecular alignment signal from the detector at different pump intensities by including both the cross-(de)focusing and anisotropic multiphoton ionization contributions. Here, we normalized the signal peaks around zero delay to unity. For a pump intensity of 1.0×10^{13} W/cm² with negligible ionization rate as shown in Fig. 1(c), the measured signal is nearly proportional to $(\langle \cos^2 \theta \rangle - 1/3)^2$ with a small correction owing to the weak cross-(de)focusing effect. As shown in Fig. 1(d), for a pump intensity of 5.0×10^{13} W/cm² with an expected ionization rate of $W_0 = 1.8\%$, a dip for the central peak around the quarter-revival time is resulted as compared to the side ones, and the second peak is relatively decreased as compared to the first one around the half-revival time.

4. Experimental results

Our experimentally measured molecular alignment signals under different pump intensities are presented in Fig. 2. When the pump intensity is lower than 10^{13} W/cm² as shown in Fig. 2(a), a homodyne signal as the conventional weak field polarization measurement was typically observed. For a pump intensity of 1.5×10^{13} W/cm², as shown in Fig. 2(b), the central peak around the quarter-revival time is relatively decreased as compared with the side ones. Around the half-revival time, the second peak is decreased in comparison with the first one. As shown in Figs. 2(c) and 2(d), such relative evolutions of the adjacent peaks become more and more pronounced with increasing pump intensities. This is consistent with our above discussion on the influence of the cross-(de)focusing effect by considering the Gaussian-shaped transverse distribution of the pump beam and the consequent intensity-dependent molecular alignment degree, which acts as additional positive or negative lens.

For a pump intensity of 5.0×10^{13} W/cm² [Fig. 2(e)], not only the cross-defocusing but also the anisotropic multiphoton ionization contributes the finally measured signal. The profile of the measured signal is almost unchanged when the pump intensity is further increased to higher than 5.0×10^{13} W/cm² as shown in Fig. 2(f). This corresponds to the formation of optical filament [24], which limits the pump intensity on the order of 5.0×10^{13} W/cm² and consequently restricts the molecular alignment as well as the ionization. To estimate the involved ionization rate W_0 , we firstly obtained a factor $\eta \sim 10$ by fitting the measured molecular alignment signals around the quarter-revival time at low pump intensities [Figs. 2(a) and 2(b), dashed curves], where only the spatial cross-(de)focusing contributions were considered due to the negligible ionizations at these intensities. Then, the measured molecular alignment signal at high pump intensity was fitted by including both the ionization and cross-(de)focusing contributions with the predetermined correction factor η . For example, at 5.0×10^{13} W/cm² as shown in Fig. 2(d), we estimated that about 3% N₂ were ionized in our

experiments. This confirms the influence of the ionization on the measured molecular alignment signal with our cross-(de)focusing assisted polarization spectroscopy technique.

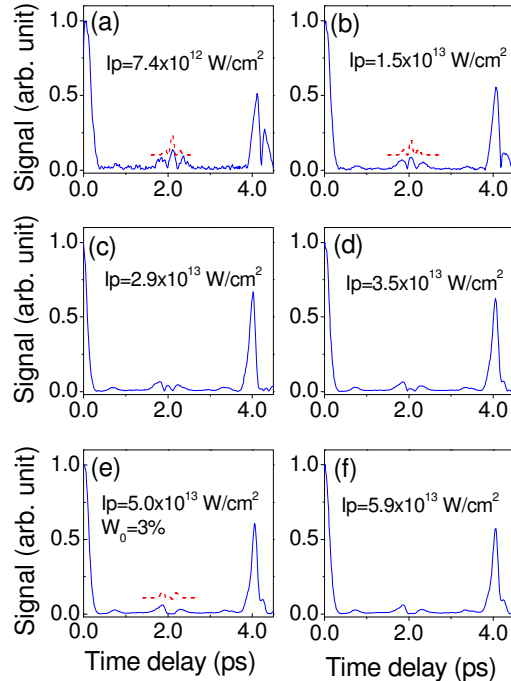


Fig. 2. Experimentally measured molecular alignment signal of N_2 under various pump intensities. The dashed curves represent the numerically fitted ones (shifted by 0.1 as compared with the measured ones).

5. Conclusion

In summary, by using a cross-(de)focusing assisted polarization spectroscopy technique, the measured molecular alignment signals are relatively increased or decreased for the parallel or perpendicular orientation revivals with increasing pump intensities. For high pump intensities, the orientation-dependent ionization of the neutral molecules during the impulsive excitation is demonstrated in the measured signal, which can be used to study the weak ionization during the molecular alignment and may be extended for filamentation in molecular gas [24] to characterize the involved ionization and clamping intensity inside the core.

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