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Femtosecond filament induced birefringence in argon and in air: Ultrafast refractive index change

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ABSTRACT

The birefringence of a linearly polarized femtosecond laser filament in gases has been previously established. In this work, we report the time-dependent refractive index measurements of the filament based upon the spectral modulation of a weak probe pulse in air and in argon gas at 1 atmospheric pressure. The polarization dependence of the refractive index modulation induced by the delayed molecular alignment and by the electronic Kerr effect is highlighted. A numerical simulation of the refractive indices, which takes into account the molecular alignment, the electronic Kerr non-linearity and the plasma, is in good agreement with the measurements.

1. Introduction

Femtosecond laser filamentation in transparent media has generated much research interest in recent years [1–6]. Current and projected applications of filamentation range from lightning control [7–9] to pollutant remote sensing [10–14], few-cycles near infrared (NIR) pulse generation [15], and single cycle THz pulse generation [16].

Filamentation is a propagation regime restricted to short laser pulses (picoseconds and below), where a low density plasma is left in the wake of the pulse [17]. Laser filamentation is observed when the peak power of intense laser pulses is higher than the critical power for self-focusing [18] in the medium. Under this condition, the Kerr self-focusing will overcome the natural diffraction and the group velocity dispersion (GVD) causing the collapse of the pulse. The main mechanism that stops the beam collapse is the diffraction by the plasma that is self-generated through multiphoton/tunnel ionization of the atoms and/or molecules in the medium [1]. As a result of the highly non-linear nature of tunneling ionization, the intensity is clamped to about 5×10^{13} W/cm^2 in the filament in air [19]. The diameter of the filament ranges typically from 50 to 100 μm [20].

Other properties of filaments were investigated recently by Béjot et al. [21]. Their experiments, performed in 3 bars of argon, showed that the Kerr effect inside a linearly polarized filament induces a large birefringence that can lead to a half-wave ($\lambda/2$) retardation. Chen et al. [22] and later Marceau et al. [23] showed that a filament in air can spatially separate the orthogonal polarizations of a probe laser beam propagating ~100 fs behind a filament generated by a pump pulse. The decomposition of an input arbitrary probe polarization into its eigen components was also highlighted prior to our work by K. Hartinger et al. [24] in aligned CO_2 molecules. As was also pointed out in Ref. [24], a gas sample under a strong laser field will act as a birefringent crystal only in the small retardation limit ($\Delta\phi \ll 2\pi$). In the non-perturbative limit, because of the non-uniform intensity profile of the laser beam, spatial effects as in Ref. [22,23] are unavoidable. The polarization separation in Ref. [22,23] was attributed to the alignment of nitrogen and oxygen molecules by the filament, which tends to guide the 0° and defocus the 90° polarization component of the laser beam. Here, 0 and 90° mean parallel and perpendicular to the linear polarization of the pump filamenting pulse. Several applications of the coherent molecular rotational wavepacket left in the wake of the filament were recently exposed, for instance, control of the onset position [25–27] or the length [28] of filamentation of a delayed pump pulse. The ~100 fs-delayed initial molecular alignment was also recently exploited by Bernstein et al. [29] to achieve energy coupling between two crossing filamenting pulses.

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Many methods to retrieve the transient refractive index changes in gases have been discussed in the literature. Several authors relied on the polarization changes of a probe pulse after passing through the birefringent gas sample [30–32]. Others used supercontinuum spectral interferometry [33–36] or cross-defocusing measurements [37]. In this work, we study the mechanism of the filament guiding/defocusing the probe's orthogonal polarizations reported in Ref. [22,23] by measuring the spectral modulation [38] of the probe pulse (due to cross-phase modulation) as a function of the delay between the pump and probe pulses. The comparison of the deduced time-dependent refractive index for both polarizations in air and in argon allows us to clearly visualize the electronic Kerr and the molecular alignment contributions in the context of atmospheric filamentation. Our discussion offers a comprehensive overview of the polarization dependence of the gas non-linearities. A numerical simulation of the refractive index modulation, which includes the electronic Kerr effect, the plasma contribution and the nonadiabatic laser-induced alignment [39–41] of O_2 and N_2, supports our results. Agreement is found between our refractive index measurements and those previously reported in the literature [35,38,42–44].

2. Theory

Throughout the paper, we assume that the propagation direction is in the z direction and the pump linear polarization is along the x direction. Modelling the filament as a cylinder of length L and of uniform clamped intensity I_{pump}, we find for the overall non-linear phase accumulated by the probe pulse through propagation in the high intensity zone of the filament [45]:

$$\phi_{\text{NL}}(r,t) = -\Delta n(r,t)\omega_0 L / c_0 \tag{1}$$

where Δn is the overall change in the refractive index caused by the electronic Kerr cross-phase modulation, the molecular alignment and the plasma; ω_0 is the laser central angular frequency; c_0 is the speed of light in vacuum; r is the radial coordinate and t is the delay time between the pump and probe pulses. The instantaneous frequency shift is given by [45]:

$$\delta\omega(t) = \frac{d}{dt}\phi_{\text{NL}}(t) = -\frac{d}{dt}\Delta n(t)\omega_0 L / c_0 \tag{2}$$

Converting this equation to the wavelength domain ($\lambda = 2\pi c_0/(n\omega)$), in the limit of small spectral shifts ($\Delta\lambda \ll \lambda_0$) and no dispersion, one obtains for the refractive index modulation:

$$\Delta\lambda(t) \approx \frac{L\lambda_0}{c_0}\frac{d}{dt}\Delta n(t) \tag{3}$$

$$\Delta n(t) \approx \frac{c_0}{L\lambda_0}\int_{-\infty}^{t} \Delta\lambda\left(t'\right)dt' \tag{4}$$

where λ_0 is the central wavelength of the laser pulse and $\Delta\lambda$ is the accumulated spectral shift (in wavelength). In the case of the pure instantaneous electronic Kerr effect in an isotropic medium, assuming that the third order susceptibility can be written as $\chi^{(3)}_{ijkl}(\omega_{\text{probe}} = \omega_{\text{probe}} + \omega_{\text{pump}} - \omega_{\text{pump}})$, it can be shown [46] that the relation $\chi^{(3)}_{xxxx} = 3\chi^{(3)}_{yyxx}$ holds. The usual non-linear refractive index n_2, defined for the self-phase modulation ($n = n_0 + n_2 I$; where I is the laser intensity) of a single linearly x-polarized pulse, is related to the third order susceptibility (in CGS units) by [47]

$$n_2 = \frac{12\pi^2}{n_0^2 c_0}\chi^{(3)}_{xxxx} \tag{5}$$

For each polarization component of the probe, the refractive index modulation caused by the electronic Kerr effect is then given by [21]

$$\Delta n_{\text{Kerr},X}(r,t) = 2n_2 I_{\text{pump}}(r,t) \tag{6}$$

$$\Delta n_{\text{Kerr},Y}(r,t) = \frac{2}{3}n_2 I_{\text{pump}}(r,t) \tag{7}$$

$$\Delta n_{\text{Kerr},X}(r,t) = 3\Delta n_{\text{Kerr},Y}(r,t) \tag{8}$$

Values for n_2 are found in the literature. In 1 bar of argon, Ref. [21] reports $n_2 = 3.2\times10^{-19}$ cm^2/W at 800 nm and $n_2 = 4.9\times10^{-19}$ cm^2/W at 400 nm, while Ref. [33] uses $n_2 = 9.8\times10^{-20}$ cm^2/W at 1055 nm. In air, a calculation from the experimental data in Ref. [48] yields 1×10^{-19} cm^2/W at 800 nm and around 40 fs. The two latter values are used here because they fit well with our experimental data.

For molecular alignment induced birefringence, in the case of linear molecules, where the two components of the polarizability tensor that are perpendicular to the molecular axis are equal and the third one (parallel) is larger ($\alpha_1 = \alpha_2 < \alpha_3$), the change of refractive indices are given by [25,49]

$$\Delta n_{\text{mol},X}(r,t) = \frac{2\pi N}{n_0}\Delta\alpha\left(\langle\cos^2\theta\rangle(r,t) - {}^1/_3\right) \tag{9}$$

$$\Delta n_{\text{mol},Y}(r,t) = -\frac{\pi N}{n_0}\Delta\alpha\left(\langle\cos^2\theta\rangle(r,t) - {}^1/_3\right) \tag{10}$$

$$\Delta n_{\text{mol},X}(r,t) = -2\Delta n_{\text{mol},Y}(r,t) \tag{11}$$

where N is the medium density, n_0 is the weak field refractive index, $\Delta\alpha$ is the polarizability anisotropy ($\Delta\alpha = \alpha_3 - \alpha_1$) and $\langle\cos^2\theta\rangle$ (r, t) is the alignment factor [39–41]. For O_2 and N_2 molecules, the polarizability anisotropies are given [27] by $\Delta\alpha_{O_2} = 1.14\times10^{-24}$ cm^3 and $\Delta\alpha_{N2} = 0.93\times10^{-24}$ cm^3, respectively.

The alignment factor $\langle\cos^2\theta\rangle$(r, t) is computed without spatial variation and as a trace operation, $\langle\cos^2\theta\rangle(t) = Tr\{\hat{\rho}(t)\cos^2\theta\}$ [41], where $\hat{\rho}(t)$is the reduced rotational density operator that obeys the quantum Liouville equation $\frac{d\hat{\rho}(t)}{dt} = \frac{i}{\hbar}[H_o + H_{\text{int}}(t), \hat{\rho}(t)] + \left(\frac{d\hat{\rho}(t)}{dt}\right)_{\text{diss}}$, H_o denotes the linear rigid rotor Hamiltonian and H_{int} is the molecule-laser (pump) interaction. The commutator part of the equation describes coherent dynamics and the bracketed last term accounts for rotational dissipative dynamics due to collisions in the gas phase [41]. Expanding the density operator in a rotational basis, we cast the quantum Liouville equation in the form of a set of coupled equations of motion of the elements of the density matrix. These are solved using a coupled differential equation solver that utilizes the fourth order Runge–Kutta method with a time step size of 93 attoseconds.

Finally, the plasma's isotropic contribution to the refractive index is given (in CGS units) by [1]

$$\Delta n_{\text{plasma}}(r,t) = -\frac{2\pi e^2 N_e(r,t)}{m_e\omega_0^2} \tag{12}$$

where e is the elementary charge, N_e is the plasma density and m_e is the electron mass.

3. Experimental setup

The experimental setup is shown in Fig. 1. Briefly, an 800 nm, 50 fs Ti-sapphire laser pulse was split into two by a 70% reflective beam splitter (BS). The reflected x-polarized (0°), 1.1 mJ, 800 nm laser pulse acted as the pump and generated a ~3 cm long single filament in a gas cell containing 1 bar of either argon or air. The transmitted pulse was frequency doubled by a Type-I 100 μm-thick KTP crystal. The resulting