Ionization-Grating-Induced Nonlinear Phase Accumulation in Spectrally Resolved Transient Birefringence Measurements at 400 nm

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We report experimental confirmation of the ionization-grating-induced transient birefringence predicted by Wahlstrand and Milchberg [Opt. Lett. **36**, 3822 (2011)] and discuss its impact on the higher-order Kerr effect interpretation by Loriot *et al.* of pump-probe transient birefringence measurements made at 800 nm [Opt. Express **17**, 13429 (2009)]. Measurement of the transient birefringence in air at 400 nm shows a negative contribution to the index of refraction at zero delay for frequencies within the pump bandwidth, the degenerate case, and no negative contribution for frequencies exceeding the pump bandwidth, the nondegenerate case. Our findings suggest that a reevaluation of the higher-order Kerr effect hypothesis of Loriot *et al.* is necessary.

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In a recent report, results from a pump-probe transient birefringence measurement were interpreted as evidence for the significance of higher-order terms in the expansion of the nonlinear index of refraction, n_{NL}, in argon, oxygen, and nitrogen [1,2]. The so-called higher-order Kerr effect (HOKE) was purported to be responsible for the saturation and subsequent sign inversion of the pump-probe birefringence at zero delay, measured in a heterodyned polarization gate geometry in Ref. [1]. Subsequently, the hypothesis of a negative HOKE contribution to the refractive index at high intensities was used to reinterpret the underlying physics of dynamic spatial replenishment of ultrashort laser pulses (filamentation) [3] as being dominated by saturation of the Kerr response over a wide range of input parameters [1,2,4]. The proposed HOKE implies that plasma-induced defocusing does not act as the primary mechanism responsible for counterbalancing Kerr-induced self focusing in a gas phase filament for short pulses in the near IR.

Wahlstrand *et al.* [5] proposed an alternative interpretation of the transient birefringence measurements of Loriot *et al.* [1], wherein the nonlinear processes in an ionization-induced grating impose a phase shift on the probe pulse through two-beam coupling, leading to an effective birefringence in the medium. A spatially stationary modulation (grating) of the ionization front in the medium originating from optical interference between degenerate pump and probe beams results in negative nonlinear phase accumulation in the probe polarization component parallel to the pump laser, with no phase accumulation in the perpendicular polarization component. Thus, for a probe pulse that is linearly polarized at an angle of 45 degrees with respect to the pump polarization (as in the Loriot measurement), a net nonlinear polarization rotation occurs while the pump and probe beams are temporally overlapped in the medium. The intensity dependence of the ionization-induced birefringence is predicted to scale as I_e^m in the multiphoton regime, where I_e is the pump intensity and m is the number of photons required to ionize the medium at the laser wavelength (with degenerate pump and probe pulses). The theory predicts that nondegenerate pump and probe beams do not result in an ionization-induced birefringence due to the lack of a stationary ionization grating. In a first approximation, this occurs when the phase velocity of the interference pattern, $|\delta|/|\mathbf{k}_e - \mathbf{k}_p|$, traverses the characteristic grating distance $d = 2\pi/|\mathbf{k}_e - \mathbf{k}_p|$ on the time scale of the laser pulse, τ. This suggests that when $\pi/\delta > \tau$, nonlinear phase accumulation can occur in the parallel probe polarization, leading to birefringence. In these expressions δ is the difference in frequency between the pump (e) and probe (p) beams and $\mathbf{k}$ is the wave vector of the respective pulses. For degenerate beams the grating is stationary and net nonlinear phase accumulation can occur; for nondegenerate beams the shifting grating will prevent net nonlinear phase accumulation through destructive interference. If the birefringence observed by Loriot *et al.* were due to the optical Kerr effect based on neutral-atomic or molecular properties, the polarization rotation would be observed regardless of the frequency of the probe beam with respect to a particular pump frequency [2,6], excepting the existence of significant dispersion in the higher-order nonlinear refractive index, which is not expected (cf. Ref. [7]).

Two previous investigations revealed no evidence for HOKE-induced saturation of the nonlinear refractive index. One was a two-color, 800 nm/400 nm pump-probe cross defocusing measurement [8]. The other used (nondegenerate) single-shot supercontinuum spectral interferometry [9] to measure the nonlinearity directly. However, no direct comparison of the two-color and degenerate

transient birefringence experiments has been performed using the transient birefringence apparatus of Loriot *et al.* Furthermore, no experiments have investigated transient birefringence with small detuning between the pump and probe pulses, where any possibility of detrimental dispersion is effectively eliminated. Here, we present spectrally resolved transient birefringence measurements in air at 400 nm for the degenerate and nondegenerate pump-probe cases using such an apparatus. We demonstrate that no sign inversion is observed where the pump and probe pulses are nondegenerate and that the intensity dependence of the observed birefringence in the degenerate case is consistent with the ionization-induced grating model.

The transient birefringence of air was measured using a pump-probe scheme similar to the one described in Ref. [1]. An 800 nm, 50 fs laser pulse was split into pump and probe pulses. The probe beam was attenuated with a neutral density filter and a half-wave plate–Glan-laser polarizer pair. A second half-wave plate was used to control the polarization of the probe after the polarizer and a 10-μm type-II BBO crystal was used to double the probe pulse. The probe beam was then reflected off of multiple dielectric mirrors coated for 400 nm to remove the 800 nm fundamental. The second wave plate and the BBO crystal were rotated to minimize the transmission through a second Glan-laser polarizer (acting as an analyzer) placed after the interaction region and before the spectrometer (USB4000, Ocean Optics) used for signal detection. The pump intensity is independently controlled using a half-wave plate and polarizer. The pump beam was doubled using a 1 mm type-I BBO crystal and reflected off of multiple 400 nm dielectric high reflectors to remove the fundamental. A second half-wave plate before the BBO crystal was used in combination with the BBO crystal to set the pump polarization at an angle of 45° with respect to the probe. The two beams were focused with an $f = 20$ cm lens into ambient air, where they crossed at a small ($\sim 3°$) angle. After the interaction region the probe beam was collimated and sent through a quarter-wave plate before the analyzer and spectrometer. The quarter-wave plate was used to generate a local oscillator for heterodyned measurements by allowing the transmission of a small portion of the probe beam through the analyzer. By rotating the quarter-wave plate $\pm 1°$ with respect to the incident probe beam polarization axis and measuring the birefringence with positive and negative local oscillator phase contributions the sign of the birefringence is obtained when the two measurements are subtracted. The heterodyne signal can be written as [1] $S_{\mathrm{het}}(t) \propto I_p(t) \otimes (\Delta n(t) + \mathcal{P})^2 - I_p(t) \otimes (\Delta n(t) - \mathcal{P})^2$, where $I_p(t)$ is the probe intensity profile; $\Delta n(t) = n_{\parallel}(t) - n_{\perp}(t)$ is the difference in the refractive index along the pump polarization axis; the refractive index along the axis perpendicular to the pump polarization, $\mathcal{P}$, is the static birefringence induced by the quarter-wave plate; and $\otimes$ is the convolution operator. The amplitude of the heterodyne birefringence signal is thus proportional to $n_{\parallel(t)} - n_{\perp}(t)$. An electronic stage in the probe path was used to control the delay between the pump and probe pulses and the quarter revivals of nitrogen and oxygen were used to calibrate the position of zero delay. The pump intensity was calculated from the pump beam parameters ($\tau_{\mathrm{pump}} \approx 75$ fs, $w_0 = 17$ μm). The heterodyned birefringence signal is spectrally resolved to provide a measurement of the phase accumulation as a function of probe frequency.

Doubling the pump and probe beams with BBO crystals of different thicknesses results in a difference in spectral bandwidth between the two pulses due to the more stringent phase-matching conditions in the thicker crystal. The FWHM bandwidth of the frequency-doubled pump is 5.2 nm, while the doubled probe FWHM is 9.5 nm. This difference in spectral bandwidth between the pump and the probe pulses allows us to investigate the effects of degeneracy and nondegeneracy on the time-resolved birefringence signal. The broader probe bandwidth provides a degenerate birefringence signal where it spectrally overlaps the pump and nondegenerate birefringence signal where there is no spectral overlap with the pump. Figure 1(a) shows the spectrally resolved transient birefringence of air measured at a pump peak intensity of 54 TW cm^{-2}. The intensity experienced by the probe will actually be smaller due to spatial averaging resulting from the noncollinear beam geometry. The effective intensity is expected to be on the order of that reported in Ref. [2] ($\sim I_{\mathrm{pump}}/1.7$), corresponding to an intensity of 32 TW cm^{-2} for the data shown in Fig. 1(a).

The data presented in Fig. 1(a) show that the birefringence undergoes a sign inversion at pump-probe degeneracy similar to that observed in the measurements presented in Ref. [1], while the birefringence in regions where there is no spectral overlap between the pump and the probe pulses (i.e., under nondegenerate conditions) exhibits no sign inversion. The sign of the nondegenerate

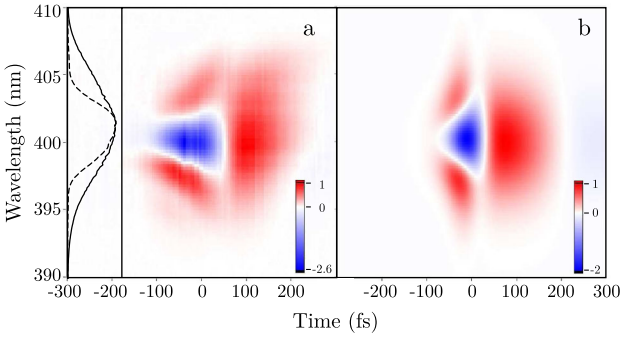

FIG. 1 (color online). (a) The heterodyned, time- and frequency-resolved birefringence of air measured at $I_{\mathrm{pump}} = 54$ TW cm^{-2}. Inset: measured pump (dashed) and probe (solid) spectra. (b) The heterodyned, time- and frequency-resolved birefringence of air calculated using Eq. (2) (see text for details).

birefringence signal is positive on both the high and low energy sides of the spectrum with respect to the nominal pump central wavelength (~ 400.7 nm), suggesting that pump photons scattered in the probe direction from the induced grating and vice versa (two-beam coupling) play a negligible role in generating the nondegenerate birefringence signal in the present experiment. The contribution of two-beam coupling to the transient birefringence has been discussed previously [2] and will not be revisited here. The positive sign of the birefringence for near-degenerate pump and probe wavelengths is inconsistent with a HOKE interpretation of the data. It is unlikely that dispersion in the nonlinear refractive index would be so large as to prevent a sign inversion at a $(\omega - \omega_0)/\omega_0 \sim \pm 0.7\%$ change in the photon energy both above and below degeneracy.

To gain insight into the effect of frequency detuning between the pump and probe pulses on the transient birefringence, we revisit the theory developed in Ref. [5]. The total electron density generated by the pump and probe fields, $N_e(\mathbf{r}, t)$, can be expressed in terms of a general ionization rate model, $W(E_{\mathrm{tot}}(t))$, that depends only on the total laser field amplitude:

$$\frac{N_e(\mathbf{r},t)}{N_0} = \int_{-\infty}^{t} dt' W(E_e(t')) + \frac{1}{2}\left(e^{i\mathbf{q}\cdots\mathbf{r}} \int_{-\infty}^{t} dt' E_p(t') W'(E_e(t')) e^{-i\delta t'} + \mathrm{c.c.}\right), \quad (1)$$

where E_e and E_p are the envelope functions of the excitation or pump laser field and the probe laser field, respectively; $\mathbf{q} = \mathbf{k}_e - \mathbf{k}_p$ is the grating wave vector; $W'(E_e)$ is the derivative of $W(E_e)$ with respect to E_e; and the carrier frequencies are retained in the expressions for the pump and probe electric fields, leading to the term $\exp(-i\delta t)$ in the grating contribution. The first integral in Eq. (1) corresponds to the smoothly varying electron density generated by the pump temporal envelope, and the second integral in Eq. (1), corresponding to the contribution leading to ionization grating-induced nonlinear phase accumulation from the pump and probe together, integrates to zero unless δ is smaller than the inverse lifetime of the ionization-induced grating formed by E_p and $W'(E_e)$. That is, if there is no spectral overlap between the pump and probe pulses, then no stationary grating is formed through ionization and there is no selective nonlinear phase accumulation in the component of the probe pulse parallel to the pump polarization. If the ionization model is assumed to be multiphoton, i.e., $W(E) = \sigma_m |E|^{2m}$, then according to Eq. (1) the amplitude of the ionization-induced change in the refractive index should scale as I_e^{m-1}, while the nonlinear source term for the transient birefringence should scale as I_e^m. For ionization of oxygen at 400 nm, a multiphoton ionization model adequately describes ionization in the pump intensity range described here, leading to the prediction of a fourth-order scaling of the negative birefringence signal with pump intensity.

To further investigate the dependence of the negative birefringence on pump laser intensity, the transient birefringence was measured as a function of laser energy and integrated over the spectrum to yield an equivalent signal to that measured in Ref. [1]. Figure 2 displays the temporally integrated birefringence amplitude over the region of sign inversion at zero delay (squares), in the region of field-free permanent alignment after zero delay (circles), and in the vicinity of the oxygen quarter revival (triangles) as a function of pump intensity, respectively. The representative error bar in Fig. 2 reflects uncertainty in the intensity calculated from measured beam parameters (estimated error of 10% pulse duration and ± 0.5 μJ) and applies to all data points. The field-free alignment and revival data are fit with exponentials of the form Ax^B, and the zero-delay signal is fit by $Ax^B + Cx$. We observe linear ($B = 1.06$) and quadratic ($B = 2.02$) behavior of the integrated oxygen revival and permanent alignment data, respectively, confirming that saturation of the alignment does not occur [10] and, more importantly, indicating that a decrease in the pump intensity due to a shift in the focal position (resulting from Kerr self focusing and/or plasma defocusing) does not significantly impact the current experiment even though measurements were performed at atmospheric pressure. The integrated zero-delay data, fit with linear and fourth-order contributions, are in agreement with the predictions of the ionization-induced birefringence model of Ref. [5].

In the experimental geometry of the present investigation, the detected signal is given by $I_{\mathrm{het}}(\omega) \propto 2\,\mathrm{Re}[E_{\mathrm{LO}}(\omega) E_s(\omega)]$ (by analogy with the time-domain expression for the total heterodyne birefringence signal in Ref. [11]), where $E_{\mathrm{LO}}(\omega)$ and $E_s(\omega)$ are the Fourier components of the local oscillator and signal fields, respectively. For carrier frequency-degenerate pump and probe pulses crossing at a small angle in the medium, the detected heterodyne birefringence signal can be approximated as

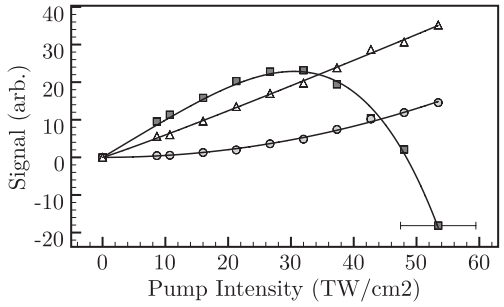

FIG. 2. The intensity dependence of the integrated transient birefringence signal at zero delay (squares), during the permanent alignment (circles), and during the oxygen quarter revival (triangles). The lines represent fits to the data (see text for details).

$$I_s(\omega) \propto \mathrm{Re}\Big[e^{i\omega t_d} E_p(\omega) iz \int_{-\infty}^{\infty} dt e^{i\omega t} \Big(-\frac{k_p n_2}{2n_0} E_p^*(t-t_d) \Big(\frac{2}{3} I_e(t) + \frac{3}{2} \int_{-\infty}^{t} dt' R(t-t') I_e(t') \Big) + 4\beta e^{iz\beta f(t)} E_e^*(t) \int_{-\infty}^{t} dt' E_p^*(t'-t_d) E_e(t') I_e(t')^{m-1} \Big) \Big], \quad (2)$$

where the first term in the Fourier transform describes the contribution from the electronic and rotational Kerr response of the medium, the second term describes the contribution from the ionization-induced grating, and z is the interaction length. Here,

$$\beta = \frac{2\pi k_p}{n_0^2} \frac{N_0 \sigma_m}{N_c}, \qquad f(t) = \int_{-\infty}^{t} d\tau I_e(t')^m,$$

and N_c represents the critical plasma density at the carrier frequency. Equation (2) retains only the leading terms in the parameter $z\beta f(\infty)$; inclusion of additional terms leads to small corrections in Eq. (2). The rotational contribution has been approximated with the response function $R(t - t_d)$ [12] rather than with a full calculation as in Ref. [1], and multiphoton ionization has been assumed. The spectrally resolved transient birefringence calculated using Eq. (2) with Gaussian input pulses ($\tau_{\mathrm{pu}} = 75$ fs, $\tau_{\mathrm{pr}} = 37$ fs) and $I_e = 32$ TW cm^{-2} is shown in Fig. 1(b). Because of the uncertainty in the value of n_2 at 400 nm (taken to be $n_2 = 5.36 \times 10^{-19}$ W cm^{-2} [13]) and in the ionization rate, the Raman-Kerr contribution has been scaled to match the experimental proportions relative to the ionization-induced grating contribution. The simulation captures the essential features of the experimental data that distinguish it from a purely Kerr-driven response, namely the positive sidebands on the leading edge of the pump pulse. Spectral and temporal integration of the pure ionization-induced birefringence as a function of intensity yielded a fourth-order power dependence, and in combination with a linear contribution (accounting for Kerr and Raman effects) agrees with the data shown in Fig. 2.

We next consider the possibility of HOKE term contributions to the measured signal and also the ramifications of the ionization-induced birefringence on the HOKE theory. In the UV (i.e., at ≤ 400 nm), plasma generation is expected to dominate over the HOKE indices [2,6], which would lead to suppression of the sign inversion of the birefringence were it due solely to the HOKE terms. Ettoumi *et al.* calculate the Kerr-canceling intensity in air at 400 nm to be $\sim$32 TW cm^{-2} based on a generalization of the Miller formulas [14], but our data show inversion well below this intensity. For comparison, inversion of the integrated temporal profile in our data occurs at a 32.1 μJ input energy, which corresponds to $I_e/1.7 = 28$ TW cm^{-2}. We note that subsequent work has cast doubt on the validity of the generalized Miller formulas [15], suggesting that the disagreement between the calculation in Ref. [6] and our data can only be qualitatively discussed. Regardless, the measurements presented here show comparable inversion of the transient birefringence as seen in Ref. [1] at 800 nm, suggesting that the sign inversion of the transient birefringence is unlikely to be a distinct indication of HOKE-dominated propagation.

To conclude, we have presented experimental results consistent with the existence of an ionization-grating-induced contribution to the transient birefringence of air measured in a pump-probe configuration similar to Ref. [1]. We measured the dependence of the sign inversion of the birefringence on pump intensity at 400 nm and showed that it scales as I_e^m, where m is the number of photons required to ionize in the multiphoton limit. We also report that the ionization-induced transient birefringence signal occurs only at degeneracy and that nondegeneracy between the pump and probe pulses results in no sign inversion of the birefringence. This is in accord with the prediction of Wahlstrand *et al.* The difference in the sign of the birefringence between degenerate and near-degenerate pump and probe wavelengths cannot be explained by the HOKE model. Our findings cast doubt on the interpretation of the transient birefringence measured by Loriot *et al.* as originating purely from higher-order terms in the Kerr nonlinearity and show that the ionization-induced grating contribution to the induced birefringence must be accounted for in order to correctly interpret the results reported in Ref. [1].

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