Thermally induced all-optical inverter and dynamic hysteresis loops in graphene oxide dispersions

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We experimentally study the temporal dynamics of amplitude-modulated laser beams propagating through a water dispersion of graphene oxide sheets in a fiber-to-fiber U-bench. Nonlinear refraction induced in the sample by thermal effects leads to both phase reversing of the transmitted signals and dynamic hysteresis in the input–output power curves. A theoretical model including beam propagation and thermal lensing dynamics reproduces the experimental findings. © 2015 Optical Society of America

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1. INTRODUCTION

There is growing interest in the nonlinear optical properties of carbon-based nanomaterials [1]. Graphene and its precursor graphene oxide (GO) have been recently proposed for novel applications in optoelectronics, biosensing, and other fields [2–4]. Dispersions of graphene and GO nanoflakes in organic and aqueous solvents exhibit broadband nonlinear optical properties. Graphene and GO dispersions exhibit optical limiting (OL) behavior under visible and infrared laser pulses in the nanosecond and picosecond regimes, at intensities of the order of GW/cm² [5–7]. In graphene dispersions under nanosecond laser pulses the mechanism responsible for OL was found to be nonlinear scattering from vapor bubbles around the graphene flakes so that the surface tension of the solvent determined the effectiveness of the OL effect [5]. In GO aqueous dispersions, saturable absorption was observed under 790 nm femtosecond laser pulses at 16 GW/cm² [8]. In the nanosecond regime, excited-state absorption was behind GO nonlinear absorption with a saturation intensity of 0.12 GW/cm² [9].

At low intensities and continuous-wave operation other nonlinear mechanisms can become significant. For example, spatial self-phase modulation was reported in graphene dispersions due to a huge broadband third-order optical nonlinearity [10]. Superluminal propagation due to reverse saturable absorption has been found in single-layer GO dispersions [11]. In the quasi-continuous regime or for long duty cycle operation, thermal effects can become apparent because heating can significantly change the local medium density. In fact, transient thermal effects have been observed up to the nanosecond regime [12].

We have recently examined in detail the phase shift in low-frequency amplitude-modulated infrared laser beams propagating through GO dispersions in water [13]. We observed small, positive phase shifts for laser powers of a few milliwatts, and large, negative phase shifts as we increased laser power. We found these phase shifts were due to thermal lens-induced OL in the GO dispersion. Thermal lensing appears when a laser beam creates a radial temperature distribution in an absorbing medium, which changes the refractive index and turns the medium into a lens. This effect was first reported about 50 years ago [14,15] but it has received renewed interest in recent years due to its potential for sensitive analysis of the physico-chemical properties of minute amounts of biological fluids and colloids [16–20].

In this work, we focus on thermally induced phase inversion in amplitude-modulated infrared beams propagating through GO dispersions. We studied the dependence of the phase reversal with the optical input power and the modulation frequency in a fiber-to-fiber U-bench setup. We observed thermally induced dynamic hysteresis in the input–output intensity plots and used them to derive the characteristic response time of our system. Finally, we developed a simple theoretical
model including beam propagation and thermal lens effect that corroborates that thermally induced nonlinear refraction in the GO dispersion is the main factor responsible for the observed phenomena.

2. THERMALLY INDUCED NONLINEAR REFRACTIVE INDEX OF GO DISPERSIONS

We used a 4 mg/ml GO water dispersion, supplied by Graphenea (Spain), which showed long-term stability. According to the manufacturer, the dispersion is made of graphene sheets with dimensions ranging from a few hundred nanometers to a few micrometers (TEM, not shown). More than 95% of the few-layer graphene sheets are monolayers, and it has a considerable degree of oxidation from C-O epoxy/ether functional groups (C1s XPS, not shown) and a C:O ratio of 1.4:1. For our experiments we diluted the commercial dispersion to concentrations ranging from 0.25 to 1 mg/ml.

We determined the linear absorption coefficient of the GO dispersions by measuring their transmittance using a continuous-wave laser beam. A quartz cuvette (Hellma, 100-QS) with a 10 mm path length contained the GO dispersion. We used a pigtailed laser diode (Thorlabs, PL975P200) operating at 977 nm. The laser was held at room temperature using a current and temperature feedback control module (Thorlabs, ITC510). We measured the power of the beam coming out of the cuvette and the power at the cuvette entrance using a power meter (Thorlabs, PM122). We obtained the linear absorption coefficient of the GO dispersions from the slope of the output/input power curves linear fit and taking into account the transmittance of the cuvette ($T_{\text{cuvette}} \approx 0.93$) (see Fig. 4(b) in [13]).

The linear absorption coefficients of the GO dispersions are $\alpha = 0.92 \ \text{cm}^{-1}, \ 1.77 \ \text{cm}^{-1}$, and 2.52 cm$^{-1}$ for concentrations 0.25 mg/ml, 0.5 mg/ml, and 1 mg/ml, respectively. These values are in agreement with values found in previous works [21].

When the GO dispersion is excited by the laser beam a temperature gradient between the center of the beam and the bulk of the sample is induced [13]. This temperature gradient causes a transverse gradient of the refractive index on the dispersion which generates a thermal lens with an inverse focal length given by [22]

$$f^{-1}_0 \equiv D_0 = \frac{(dn/dT)P(1 - e^{-\alpha L})}{\kappa \pi w_0^2}, \quad (1)$$

where $dn/dT, \kappa, \alpha$, and $L$ are the thermo-optic coefficient, the thermal conductivity, the linear absorption coefficient, and the thickness of the medium, respectively; $w_0$ is the beam radius; and $P$ is the beam power. This expression was first derived by Gordon et al. [22] assuming a perfect thin lens [22–24]. Later on, the aberrant nature of the thermal lens was analyzed by Sheldon et al. using a diffraction integral approach [15]. Bialkowski and Chartier obtained equivalent results with a simpler method based on the calculation of cumulative electric-field phase shifts produced by a series of Gaussian refractive-index perturbations [25]. A complete thermal lens model for characterization of solid materials has recently been developed by Malacarne et al. [26].

In our system, the focal length predicted with Eq. (1) is of the order of a few meters. This estimation was done using the thermal conductivity and the thermo-optic coefficient of water [27], i.e., $\kappa = 0.6 \ \text{W/(m K)}$ and $dn/dT = -0.45 \times 10^{-4} \ \text{K}^{-1}$, a beam radius $w_0 = 1.55 \ \text{mm}$, the experimentally measured linear absorption coefficient $\alpha$, and a path length of $L = 1 \ \text{cm}$. The GO dispersion temperature change $\Delta T$ is about one kelvin, and its thermal nonlinear refractive index is $n_2 = (dn/dT) w_0^2 (1 - e^{-\alpha L})/(2\kappa L) \approx 10^{-6} \ \text{cm}^2/\text{W}$ for the GO concentrations used in this work.

3. ANALYSIS OF THE BEAM TRANSMISSION THROUGH THE SYSTEM

The thermally induced lens in the GO dispersion has a power-dependent focal length [see Eq. (1)] which renders it as a power controller. In our previous work [13], we have shown OL when placing the GO dispersion in a fiber-to-fiber U-bench. Here we study the beam propagation through the fiber-based experimental system depicted in Fig. 1. The quartz cuvette with the GO dispersion was placed at the center of a fiber-to-fiber U-bench (Thorlabs, FBC-1550-APC). The U-bench had a free-space length of 59.8 mm and two glued AR-coated aspheric lenses for light collimation and coupling. According to the manufacturer, the typical collimated beam diameter is 3.1 mm (at 1550 nm). The pigtailed laser beam was connected to a single-mode optical fiber and launched through the collimating lens L1 into the cuvette with the GO dispersion. The transmitted beam was coupled into another single-mode fiber by a focusing lens L2. The output beam power was measured by placing a power meter sensor at the distal end of the single-mode optical fiber connected to the U-bench, as shown in Fig. 1. The power of the beam impinging on the cuvette (input power) was measured by replacing the cuvette with the power meter sensor in the U-bench.

In Fig. 2 we plot the output–input power curves for dispersions with different GO concentrations. We also plot the water output–input power curve for comparison purposes. We observe that the output power tends to saturate with increasing input power. A maximum in the input–output power curves is reached, above which the output power decreases for increasing input power. This maximum depends on the GO concentration. The larger the GO concentration, the smaller the input power needed to observed this change in behavior. The threshold input power is 33 mW for 1 mg/ml, and 37 mW for 0.5 mg/ml [see Fig. 2(b)].

![Fig. 1. Experimental setup. C, cuvette with the GO dispersion; DFB LD, distributed feedback laser diode at 977 nm; LD TEC, laser diode temperature and current controller; PM, power meter sensor; L1, collimating lens; L2, focusing lens; green lines, single-mode optical fibers with FC/APC connectors.](image-url)
In a previous work [13] we theoretically studied the propagation of a beam through a GO dispersion in a fiber-to-fiber U-bench. We used the ray transfer matrices formalism to describe the propagation of a Gaussian beam through our system [28] (in the paraxial wave approximation). The cuvette was placed at the center of the U-bench being the free-space distance behind it around $d = 25 \text{ mm}$. We considered that a quasi-planar wavefront Gaussian beam with wavelength $\lambda = 977 \text{ nm}$, spot radius $w_0 \approx 1.55 \text{ mm}$, and radius of curvature $\approx \infty$ emerges from L1 and impinges into the cuvette. As we mentioned before, this beam is partially absorbed by the GO dispersion, which produces a thermal lens whose focal length is $f$.

From the beam exit to the L2 entrance the beam only propagates a distance $d \ll f$ in free space. Therefore we ignore diffraction/propagation effects from the cuvette’s output to lens L2. Thus, at the L2 entrance the beam radius is $w_0$ and the radius of curvature is $f$. L2 focuses the beam into a single-mode fiber, whose entrance plane is located at $d_L = 11 \text{ mm}$. This value was estimated by using the single-mode fiber numerical aperture (NA = 0.14) and the typical spot radius $w_0$, i.e., $NA \approx w_0 / d_L$.

The maximum of function $T_{MM}$ occurs at $w = w_f$ which satisfies

$$T_{MM} = \frac{\alpha}{w^2 + w_f^2},$$

where $w_f$ is the radius of the fiber mode. $T_{MM}$ represents the transmittance of the optical system, i.e., $T_{MM}P$, is shown in Fig. 3 as a function of the input power $P$. A maximum output power is reached for an input power close to $40 \text{ mW}$. Above this input power, output power decreases for increasing input power. This negative input–output characteristic will allow us to obtain optical signal inversion, i.e., an amplitude-modulated beam with an input power greater than $40 \text{ mW}$ could reverse its phase after transmission through the system with a 0.5 mg/ml GO dispersion. The threshold input power ($\approx 40 \text{ mW}$) leads to a beam spot radius $w_L$ close to $6 \mu m$.

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$$w_L = w_0d_L\sqrt{\left(\frac{1}{f}\right)^2 + \left(\frac{\lambda}{\pi w_0^2}\right)^2}.$$
characteristics of our setup is $P_{out} = e^{-\alpha T_{cuvette}} T^0 T^{MM} P_0$, where $T^0 = 0.5$ is the measured loss of our optical system. In Fig. 2 we plot the dependence of the theoretical output power $P_{out}$ (solid lines) with input power, and we observe a good agreement with the experimental results.

4. REVERSED-PHASE WAVEFORM RESULTS

To test the previous prediction about reversed-phase waveform phenomenon, we analyzed the propagation of sinusoidally amplitude-modulated beams through the experimental system depicted in Fig. 4 for a GO dispersion with a concentration of 0.5 mg/ml. The laser injection current was modulated with a function generator (Agilent, 33220A). The transmitted laser signal was measured with a switchable-gain amplified InGaAs photodetector placed at the distal end of the single-mode optical fiber connected to the U-bench (Thorlabs, PDA10CS-EC). An inline variable optical attenuator was used to avoid saturation of the photodetector. We used a digital oscilloscope to record the photodetector signal averaged over 16 oscilloscope traces (Agilent, DSO9104A). We measured the beam transmitted through the system with and without the GO dispersion cuvette. In both measurements, with and without the GO cuvette, we used the function generator signal that modulates the laser injection current as the oscilloscope trigger. Thus, the function generator signal acts as a shared reference to compute the laser injection current as the oscilloscope trigger. Consequently, the cuvette, we used the function generator signal that modulates the phase delay induced by the dispersion. For these experiments, a sinusoidal current was applied so that the beam power is modulated as a function of time (TM ventilation). The transmitted laser signals for different modulation frequencies are shown for different values of the modulation frequency $f_m$. The average input power is 80 mW and the modulation amplitude is 10%.

We performed the correlation between input and output signals to analyze the behavior of the phase shift versus the modulation frequency $f_m$ and the average input power. Figure 6(a) shows the experimental phase shift dependence with modulation frequency (symbols) for a laser average input power of 80 mW. We observe that for modulation frequencies smaller than 0.1 Hz the system acts as an optical inverter while the phase shift almost vanishes at frequencies larger than 100 Hz. The behavior of the phase shift with average input power is shown in Fig. 6(b) for a modulation frequency of $f_m = 0.2$ Hz. Reversed phase output waveforms are found for input powers above 50 mW. Large signal-reversing shifts are obtained at high input power and for low frequency beams, which indicates that laser-induced heating of the GO dispersion is the mechanism underlying this phenomenon.

A. Theoretical Model: Phase Reversal

To theoretically reproduce this effect we consider that the modulation of the beam power produces a time-varying

![Fig. 4. Experimental setup. C, cuvette with the GO dispersion; DFB LD, distributed feedback laser diode at 977 nm; LD TEC, laser diode temperature and current controller; FG, function generator; OC, digital oscilloscope; VOA, variable optical attenuator; PD, photodetector; L1, collimating lens; L2, focusing lens. Green lines, single-mode optical fibers with FC/APC connectors. Gray thick lines, coaxial cables with BNC connectors.](image-url)

![Fig. 5. Experimental time evolution of the sinusoidal amplitude-modulated signals. The input (red line) and output (black line) signals are shown for different values of the modulation frequency $f_m$. The average input power is 80 mW and the modulation amplitude is 10%.](image-url)

![Fig. 6. Experimental (symbols) and theoretical (line) phase shift versus: (a) modulation frequency $f_m$ for a laser average input power of 80 mW; (b) input power for a modulation frequency of 0.2 Hz. In both cases the modulation amplitude is 10%. For the theoretical curve we use the following parameters: $\kappa = 0.6$ W/(m K), $dn/dT = -0.45 \times 10^{-4}$ K$^{-1}$, $w_0 = 1.55$ mm, $\alpha = 1.77$ cm$^{-1}$, $L = 1$ cm, $d_L = 11$ mm, $\lambda = 977$ nm, $\tau = 0.2$ s, and $\omega_f = 3.1$ $\mu$m.](image-url)
temperature spatial profile on the sample, which in turn results in an oscillating focal length of the thermal lens induced in the dispersion [13]. Let us assume the cuvette as an infinite medium since the beam radius, 1.6 mm, is smaller than the cuvette transverse size. This approximation allows us to achieve analytical results by solving the heat equation in the Fourier domain. Then, the temperature spatial transverse profile, $\Delta T$, will be obtained by computing the inverse Fourier transform (Hankel transform). The heat equation in the Fourier domain reads

$$\frac{\partial \Delta T}{\partial t_d} = \frac{P(1 - e^{-al})}{kL} e^{i\xi q} - (2\pi)^2 q^2 \Delta T,$$

where $\Delta T$ is the spatial Fourier transform of $\Delta T$, and $q$ is the wavenumber normalized to the beam radius ($1/w_0$). The time variable $t_d = t/\tau$ has been normalized to the thermal lens buildup time $\tau = C_p w_0^2/k$, $C_p$ being the heat capacity per unit volume ($C$ is the heat capacity per unit mass and $\rho$ is the medium density). This characteristic time is usually in the range between milliseconds and tenths of second. The model assumes a small beam divergence angle so that the beam radius basically does not change along the sample. In fact, the Rayleigh length is of the order of meters, much greater than the sample thickness. In our experiments, the absorption length is comparable to the sample thickness so an axial dependence on the temperature profile could be expected. This variation has been addressed in detail in [26] when studying the laser-induced wavefront distortion. They found that for moderate absorption media the temperature axial profile decays following the attenuation of the beam power, i.e., $\exp(-az)$. Thus, the optical path length is proportional to the effective length ($1 - \exp(-(al))/a$. In our model we directly include this effective length in the source term to roughly account for sample absorption.

Let us assume that the temperature change is forced to oscillate at the same frequency as the beam power:

$$\Delta T = \Delta T_0 + \Delta T_c \cos(\omega_m t_d) + \Delta T_s \sin(\omega_m t_d),$$

where $\omega_m = 2\pi f_m \tau$ is the normalized (dimensionless) modulation angular frequency. The steady-state value ($\Delta T_0$) and the amplitudes of the temperature change oscillation ($\Delta T_c$, $\Delta T_s$) are easily obtained from the heat equation (4):

$$\Delta T_0 = \frac{P_0(1 - e^{-al})}{kL} e^{i\xi q} \frac{1}{(2\pi)^2 q^2},$$

$$\Delta T_c = \frac{P_m(1 - e^{-al})}{kL} e^{i\xi q} \frac{(2\pi)^2 q^2}{(2\pi)^2 q^2 + \omega_m^2},$$

$$\Delta T_s = \frac{P_m(1 - e^{-al})}{kL} e^{i\xi q} \frac{\omega_m}{(2\pi)^2 q^2 + \omega_m^2}.$$

We numerically compute the inverse Fourier transform of Eqs. (5)–(7) by means of a Hankel transform,

$$\Delta T_l = 2\pi \int_0^\infty dq \Delta T_l / f_0(2\pi q_r d) q, \quad (l = 0, c, s),$$

where $f_0$ is the Bessel function of the first kind of order zero and $r_d = r/w_0$ is the dimensionless radius normalized to the beam radius $w_0$. In order to obtain analytical results we resort to use the parabolic approximation for the temperature profile. Thus, we approximate the spatial profile of $\Delta T_l$ in the proximity of the beam center by retaining the second-order term of the power series expansion of $f_0$. In our model we are not taking into account the aberrant nature of the thermal lens [15]. This effect could slightly change the coupling efficiency of the aberrated beam to the optical fiber but it does not affect the dynamical behavior analyzed in this work, as we will show below. Then, the temperature change reads

$$\Delta T_0 = -r_d^2 \frac{P_0(1 - e^{-al})}{2\pi kL},$$

for the steady state, whereas the oscillation amplitude of the temperature change $\Delta T_c$ and $\Delta T_s$ are given in terms of the exponential integral function $E_i(x) = \int_x^\infty dy y^{-i}$:

$$\Delta T_c = -r_d^2 \frac{P_m(1 - e^{-al})}{4\pi kL} \left[-i \frac{\omega_m}{8} e^{-\omega_m} E_i \left(\frac{\omega_m}{8}\right) + 2\right],$$

$$\Delta T_s = -r_d^2 \frac{P_m(1 - e^{-al})}{4\pi kL} \left[\frac{\omega_m}{8} e^{-\omega_m} E_i \left(\frac{\omega_m}{8}\right) + 2\right].$$

Then, the dioptric power (inverse focal length) of the thermal lens induced by the laser beam oscillates as follows,

$$\frac{1}{f} = D - 2L \frac{d}{dT} \frac{\Delta T}{w_0^2 d},$$

where the steady-state value $D_0 = f_0^{-1}$ corresponds to the well-known expression given in Eq. (1). $D_c$ and $D_s$ represent the amplitude of the thermal lens dioptric power oscillation. With the value of $f$ calculated from Eq. (12) we compute the beam radius $w_{l_d}$ at the entrance plane of the output single-mode fiber of the U-bench [Eq. (2)] and the corresponding coupling efficiency $T_{MM}$ [Eq. (3)]. Then, we finally compute the output laser power $P_{out} = e^{-al} T_{cuvette} T_{MM} P_{in}$.

As an example, in Fig. 7 we plot the time evolution of these magnitudes for an input power with $P_0 = 80$ mW, $P_m = 0.1 P_0$, and $\omega_m = 0.25$. The input power is plotted on the right axis for comparison purpose (red dashed line). In Fig. 7(a) we see that the inverse focal length $D$ (solid line) exhibits phase inversion with respect to the input power (dashed line). This produces the same behavior in $T_{MM}$ [see Fig. 7(c)], which finally leads to the reversal of the output waveform phase [see Fig. 7(d)]. This behavior agrees with the experimental findings shown in Fig. 5 (upper left panel) where low-modulation frequency transmitted signals reverse their phase for an input power of $P_0 = 80$ mW, and $P_m = 0.1 P_0$. Reversed phase waveforms have been found in erbium-doped materials due to negative nonlinear absorption effect, which causes transmittance to decreases as the laser intensity increases [30–32]. Optical bistability derived from negative nonlinear absorption has been also found in the same system [33,34]. In our case, as was shown in Fig. 3, reversed-phase phenomenon arises due to the interplay between laser-induced thermal lensing in the GO dispersion, and coupling efficiency of the
5. DYNAMIC HYSTERESIS RESULTS

With the aim of studying the temporal dynamics of our system, we look for the presence of dynamic hysteresis in the input–output power curves. Thermally induced dynamic hysteresis has been found in the turn-on and turn-off of vertical-cavity surface-emitting lasers [35]. The hysteresis cycles depend on the current modulation frequency and the substrate temperature. In our case, we linearly change the laser injection current by means of a triangular waveform with period $T$, $\Lambda_T(t)$. The injection current was varied from a value nearly above laser threshold ($\approx 70 \text{ mA}$) to a value close to the maximum allowed by the current controller ($\approx 440 \text{ mA}$) being half of the period $T$ the time to go from the minimum to the maximum value of the current. This leads to a change in the input power from $P_1 \approx 7 \text{ mW}$ to $P_2 \approx 95 \text{ mW}$. Figure 8 shows the output–input power curves for different values of the period $T$. We see that rate-dependent loops appear. In particular, for long periods, transmitted power decreases as input power increases above a threshold value around $40 \text{ mW}$, in agreement with the steady-state results. The threshold power above which a negative input–output characteristic appears increases when decreasing the modulation period $T$. Thus, the thermally induced hysteresis observed depends on the interplay between two characteristic times: the injection current modulation period $T$ and the typical thermal lens formation time. If the power modulation is too slow, a steady-state thermal lens is generated in the dispersion before the injection current changes again. In this case, hysteresis does not occur. On the other hand, if the current modulation is too fast, the thermal lens induced in the GO dispersion cannot follow the input power changes and an average optical power of around $50 \text{ mW}$ seems to be impinging on the dispersion. By inspecting Fig. 3 we see that an input power of $50 \text{ mW}$ leads to $T_{\text{MM}} = 0.5$ so a total transmittance of around $4\%$ is obtained, which agrees with the one observed in bottom right panel of Fig. 8. Thus, a thermal lens corresponding to that average power is induced in the laser beam to the output fiber of the U-bench. Therefore, the thermally induced nonlinear refractive index of the GO dispersion leads to an all-optical inverter.

We compute the dependence of the phase shift between the output and input beams with both the modulation frequency and the input power. In Fig. 6 (solid lines) we plot the simulated phase shift versus the modulation frequency $f_m$ (a), and the input power (b). These curves have been obtained varying the thermal lens formation time $\tau$ to fit the experimental ones, obtaining $\tau = 0.2 \text{ s}$. A good agreement can be seen. Thus, reversed-phase transmitted signals are only observed for modulation frequencies slow enough so that the variation of the thermal-lens focal length can occur [see Fig. 6(a)]. Furthermore, an input power above threshold ($40 \text{ mW}$) is needed for phase reversal in agreement with the results in Fig. 3.

An analytical expression of the phase shift $\Phi$ can be easily obtained taking into account that the oscillation amplitude of the thermal lens dioptric power is much smaller than $D_0$,

$$\tan \Phi = \frac{2w_0^2d_L^2(w_f^2 - w_{th}^2)D_0D_p}{w_{th}^2(w_f^2 - w_{f, th}^2)D_0D_p + 2w_0^2d_L^2(w_f^2 - w_{th}^2)D_0D_p},$$

(13)

where $w_{th} = w_0d_L\sqrt{D_0^2 + (\lambda/(nw_0^2))^2}$ is the steady-state beam radius. The phase shift obtained through Eq. (13) matches the previous calculated curves shown in Fig. 6 (solid lines).
dispersion and neither hysteresis nor power controller is achieved.

We analyzed the size of the hysteresis loops as a function of the modulation period \(T\). We compute the area inside the cycle for different values of \(T\). Figure 9 shows the result. The size of the hysteresis loops achieves a maximum for a modulation period close to \(T \approx 0.2\;\text{s}\), which agrees with the thermal lens formation time that we used to fit our experimental reversed-period close to the hysteresis loops achieves a maximum for a modulation period \(P\) [Eq. (4)]. The modulation of the input power can be written as

\[
\frac{\Delta T}{T_d} = -\frac{2P_m(1 - e^{-\alpha t})}{\kappa r^3 L} - \frac{(n-1)(n-1/2)}{n^2} \frac{2\pi q}{T_d} e^{\pi i q t} E_q + \frac{4\pi q}{T_d} e^{-\pi i q t} E_q \quad (16)
\]

\[
\Delta T_{cm} = \frac{8P_m}{\kappa r^2 L n^2} \left(1 - e^{-\alpha t}\right) \left(1 - \frac{(n-1)(n-1/2)}{n^2}\right) \frac{2\pi q}{T_d} e^{\pi i q t} E_q + \frac{4\pi q}{T_d} e^{-\pi i q t} E_q \quad (17)
\]

We numerically compute the inverse Fourier transform of \(\Delta T(l = 0, sn, cn)\) by means of the Hankel transform and consider the parabolic approximation. The steady-state value of the temperature change \(\Delta T_0\) is given by Eq. (9) whereas the oscillation amplitudes of the temperature change remain:

\[
\Delta T_{sn} = -\frac{r_d}{\kappa r^3 L} \left(1 - e^{-\alpha t}\right) \left(1 - \frac{(n-1)(n-1/2)}{n^2}\right) \frac{2\pi q}{T_d} e^{\pi i q t} E_q + \frac{4\pi q}{T_d} e^{-\pi i q t} E_q \quad (18)
\]

\[
\Delta T_{cn} = \frac{r_d}{\kappa r^3 L} \left(1 - e^{-\alpha t}\right) \left(1 - \frac{(n-1)(n-1/2)}{n^2}\right) \frac{2\pi q}{T_d} e^{\pi i q t} E_q + \frac{4\pi q}{T_d} e^{-\pi i q t} E_q \quad (19)
\]

The temporal dynamics of the dioptric power (inverse focal length) of the thermal lens induced by the laser beam will be as follows,

\[
1/f \equiv D = D_0 + \sum_{n=1}^{\infty} 2L \frac{dn}{w_0^2} \left(-\frac{\Delta T_{sn}}{T_d}\right) \frac{\sin \left(\frac{2\pi n t d}{T_d}\right)}{w_0^2} + \sum_{n=1}^{\infty} 2L \frac{dn}{w_0^2} \left(-\frac{\Delta T_{cn}}{T_d}\right) \frac{\cos \left(\frac{2\pi n t d}{T_d}\right)}{w_0^2}, \quad (20)
\]

where the steady-state value \(D_0\) is given by Eq. (1). Following the same procedure as in Section 4A, once the value of \(f\) has been obtained using Eq. (20), we compute the beam radius \(w_1\), the coupling efficiency \(T_{MM}\), and the output laser power \(P_{out}\). We theoretically found different hysteresis loops depending on the value of the modulation period, in agreement with the experimental findings. In particular, we calculate the dependence of the size of the hysteresis loops with the modulation period \(T\) by considering the sum of the first 11 terms \((n = 1, \ldots, 11)\) in Eq. (20). The simulated curve is shown in Fig. 9 (solid line) and shows a good agreement with the experimental results. We used the same parameters as the ones used in Fig. 6.

In summary, the dynamics of thermal lens focal length induced in the water GO dispersion and the mode-matching coupling between the output cuvette beam and the single-mode fiber explain the experimental behavior found in our system.

6. CONCLUSIONS

We have experimentally and theoretically investigated the propagation of amplitude-modulated infrared laser beams through GO water dispersions in a fiber-to-fiber U-bench setup. We have experimentally characterized and modeled the laser beam transmission through the system for different GO concentrations. At high concentrations and above a threshold input power, output power decreases for increasing input
power. Thus, sinusoidal amplitude-modulated transmitted signals undergo phase inversion for low modulation frequencies. This all-optical signal inversion is explained by thermal lens effects, that is, thermally induced refractive index nonlinearities in the GO water dispersions.

We have also examined the dynamic hysteresis loops in the input–output power plots when using a laser current ramp. This hysteresis depends on the interplay between the characteristic thermal lens buildup time and the external injection current modulation period. The maximum size of the hysteresis loops occurs when these times match. The hysteresis loops can be used to determine the characteristic time of the thermal lens and therefore the frequencies at which phase inversion can be observed.

Analytical expressions for the thermal lensing temporal dynamics in the GO dispersion have been obtained by solving the heat equation in Fourier space in the parabolic approximation. Our theoretical model reveals that both the phase inversion and dynamic hysteresis loops arise from the temporal dynamics of the thermal lens induced in the GO dispersion, and the matching between the beam mode coming out the cuvette and the beam mode propagating through the output fiber of the U-bench.

The findings of this work can be extended to other wavelengths and other carbon-based dispersions due to their ultrawideband response and strong light absorption. We envision that the phenomena analyzed in this work may find application in the context of optical limiting, thermal spectroscopy, and optofluidics, to realize fluid-defined optical functions [36,37].

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