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Mid-infrared all-optical modulators based on an acetylene-filled hollow-core fiber

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Abstract

We report all-optical mid-infrared phase and intensity modulators based on the photo-thermal effect in an acetylene-filled anti-resonant hollow-core fiber. Optical absorption of the control beam promotes the gas molecules to a higher energy level, which induces localized heating through non-radiative relaxation and modulates the refractive index of the gas material and hence the accumulated phase of the signal beam propagating through the hollow-core fiber. By modulating the intensity of the control beam, the phase of the signal beam is modulated accordingly. By use of a 1.53 μ m near-infrared control beam, all-optical phase modulator placed in one arm of a Mach-Zehnder interferometer, intensity modulation with on-off ratio of 25 dB is achieved. The gas-filled hollow-core-fiber modulators could operate over an ultra-broad wavelength band from near- to mid-infrared and have promising application in mid-infrared photonic systems.

Keywords: Optical phase modulator, Mid-infrared, Hollow-core fiber, Mach-Zehnder interferometer, Photothermal effect

Introduction

Photonics has attracted great research interest over the last decade for its prospects in the creation of functional devices both for near-infrared (NIR) and mid-infrared (MIR) wavelengths¹. Optical modulators are key elements of photonic circuits that enable signal switching and routing, data encoding, phase-sensitive detection, and spectroscopic interrogation². For NIR modulators, combining with optical platforms like fibers, grating and resonators, various types of materials, especially two-dimensional (2D) materials, have

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²Photonics Research Center, The Hong Kong Polytechnic University Shenzhen Research Institute, Shenzhen, China spurred a variety of researches recently³⁻⁵. Tan et al. reviewed the properties of 2D materials and introduced the design, fabrication and application of these 2D materialbased optical devices³. Zheng et al. reported a four-layer phosphorene-decorated microfiber for all-optical modulation at telecom band. They also demonstrated that the mode-locked fiber laser can be used as control light to modulate on/off continuous wave signal light with modulation depth of 4.7⁴. Wang et al. reported an all-optical phase modulator using phosphorene-deposited microfiber. A maximal phase shift of 8π rad is obtained with control light of 290 mW at 1550 nm. Such a phase modulator is further demonstrated as an information loader with modulation depth of 17 dB and response time of 2.5 ms⁵.

The motivation for investigating modulator wavelengths further into MIR comes from the applications that are possible for photonics and sensing in this range. MIR

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modulators has a wealth of applications, including actively Q-switched laser generation, environmental pollution monitoring, chemical and bio-sensing, industrial processes control, multispectral thermal imaging, and medical diagnostics of early disease, etc^{1,2,6}. To date, most of the MIR modulators are established on waveguide-integrated or free-space platforms⁷⁻⁸. On-chip integrated devices have attracted great attention due to it s flexible waveguide geometry and complementary metal oxide semiconductor compatibility. Various waveguide- integrated modulators have been demonstrated, including those based on electrooptic⁹, thermo-optic¹⁰, free carrier plasma dispersion¹¹, and electro-absorption effects¹². Most waveguide-integrated modulators are based on the silicon-on-insulator¹³ or silicon-on-lithium-niobate¹⁴ platform, which are mainly used in the NIR region due to stronger material absorption at longer wavelengths¹⁵. For free-space modulators, metamaterial¹⁶, hybrid¹⁷ and pattern structures¹⁸ are commonly used to enhance light-matter interaction to achieve higher modulation efficiency. Most of the MIR modulators are electrically driven with external electronics applied directly on the modulation devices⁶.

Here we report a novel method to achieve MIR phase modulation by exploiting photo-thermal (PT) effect in an acetylene (C_2H_2) -filled anti-resonant hollow-core fiber (AR-HCF). PT effect in gas-filled HCFs has been exploited for ultra-sensitive gas detection¹⁹⁻²¹. Gas absorption of a pump beam generates heat, which modulates the phase of an NIR probe beam propagating through the fiber. Interferometric detection of the phase modulation allows the retrieval of gas concentration. The long interaction distance and near perfect overlap of the pump and probe fields with gas material in the HCF significantly enhance light-gas interaction, which enables larger phase modulation and hence better gas sensitivity over free-space systems^{22,23}. In the present work, we extend the application scope of PT effect in the gas-filled HCFs to MIR phase modulators. Different from the voltage-driven waveguide and free-space modulators, the PT MIR phase modulators are driven optically by a cost-effective control laser in NIR telecom band. By placing the phase modulator (PM) in one arm of a Mach-Zehnder interferometer (MZI), MIR intensity modulation (IM) is further demonstrated. The AR-HCFs intrinsically have broadband transmission, which coupled with the narrow absorption lines of gas materials, makes it possible to develop ultra-broad band all-optical modulation devices from NIR to MIR.

Results

Methods and experiments

The basics of the phase modulator are shown in Fig. 1. It



uses an AR-HCF with cross-section shown in the inset of the figure²⁴. The AR-HCF consists of a single ring of six non-touching silica capillaries with diameter of ~36 μ m and thickness of ~0.76 μ m, giving an inscribed inner hollow-core diameter of ~70 μ m. The outer diameter of the fiber is 250 μ m. The AR-HCF has a low transmission loss of less than 0.5 dB/m from 1.55 μ m to 3.35 μ m, which is measured using a cut-back method²⁵. The transmission window of the state-of-the-art AR-HCFs can be further extended to longer MIR wavelengths²⁶.

The AR-HCF is filled with pure C_2H_2 at different gas pressure. Optical absorption of the control beam promotes the gas molecules to a higher energy level, which induces localized heating through non-radiative relaxation and modulates the refractive index (RI) of the gas material and hence the accumulated phase of the signal beam propagating through the AR-HCF. By placing the phase modulator in one arm of an MZI, an optically controlled intensity modulator can also be realized.

The experimental set-up is depicted in Fig. 2. It may be regarded as comprising of three modules: the optical driver, the PM, and the MZI. The MZI is used to characterize the PM, as well as to form, in combination with the PM, an intensity modulator (IM). In the optical driver, a distributed feedback (DFB) laser is used as the control light source and its wavelength is tuned to the P(9)absorption line of C₂H₂ at 1530.371 nm. The intensity of the control beam is modulated by an acoustic optical modulator (AOM) and amplified by an erbium-doped fiber amplifier (EDFA). A wavelength-division multiplexer (WDM) with narrow transmission bands is used to filter out the amplified spontaneous emission noise from the EDFA. The WDM is also used to couple an NIR signal beam into the system, as will be described in later section. An optical collimator (OC) is used to collimate the control beam, which is coupled into the HCF via a dichroic mirror (DM reflectivity: 99%@1.53 µm, 95%@1.55 μm: transmittance: 80%(a)3.35 µm) and a convex lens (f = 25 mm).

The gas-filled HCF is the core element of the PM. The

two ends of the HCF are sealed inside two gas chambers with CaF_2 windows for light coupling. A gap of ~1 mm is introduced between the CaF₂ windows and the HCF, which enables gas-filling into the HCF at different gas pressure. It should point out that, for practical applications, the HCF will be sealed with high tightness once the gas type, pressure and concentration are confirmed, therefore no external gas chamber, pump, and gas control system is required. A 3.35 µm interband cascade laser (ICL) is used as the MIR signal source to characterize the phase modulator. The insertion loss of the PM is \sim 3 dB, mainly caused by the signal beam-HCF coupling efficiency, this can be further optimized by using the MIR single-mode fiber-HCF coupling method²¹. The MIR signal beam is split into two by the DM. One beam passes through the PM while the other acts as a reference beam, which are spatially combined with a spatial beam splitter (BS) with a splitting ratio of 50:50 and further focused into a photodiode. A filter is used to block the residual control beam (around 1530 nm) from the AR-HCF. A servo-loop with a piezo-mounted mirror is used to stabilize the MZI to a particular operating point.

The MZI, with the PM placed in one arm, can also be used as an IM. To balance the optical power in the two arms, two irises are used, one in each arm. The insertion loss of the IM is ~6.9 dB at 3.35 μ m and ~13 dB at 1.55 μ m. The higher loss at NIR band is attributed to the coupling loss into HCF (~3 dB), the natural loss of the BS (3 dB), and the DM that has low transmittance at 1.55 μ m. This loss may be reduced by optimizing the coupling efficiency into the HCF, and using a more suitable DM and iris to match the power distribution of the two MZI arms.

MIR phase modulator performance

The performance of a phase modulator made of a 19-cmlong AR-HCF filled with C_2H_2 is evaluated using the setup in Fig. 2 with the MZI stabilized at quadrature point. Fig. 3a shows the DC-output from the MIR photodetector without servo-control. As shown in Fig. 3b, the MZI output with servo-locking around the quadrature point is used to estimate the long-term stability of the MZI. The output amplitude reaches a constant level immediately after the servo stabilization is applied, and the standard deviation of the phase fluctuation is calculated to be 0.04° during the monitoring time of 25 min, verifying the stability of the MZI.

Fig. 4a shows a typical MZI output waveform with the control beam modulated sinusoidally in intensity at 3 kHz. The control power delivered to the AR-HCF is 650 mW and the gas pressure is 3.3 bar. It should be noted that the control power is measured after the HCF output, which is







approximately equal to the power inside the HCF due to the low transmission loss at ~1530 nm (<0.1 dB with 19 cm-long HCF). The phase modulation amplitude is read out by using a method described in Ref. 27, where a timedomain method is demonstrated in which the dynamic phase modulation index applied to an interferometer is directly measured from the MZI waveform. For the waveform shown in Fig. 4a, T represents one control modulation period, and the phase modulation is calculated based on the waveform within half of a modulation period (with highlighted curve in Fig. 4a, from V₁ to V₂), to be 2.14π rad. With this method, the frequency response of the MIR phase modulator is determined and shown in Fig. 4b. The phase modulation amplitude reduces with increasing modulation frequency. With reference to the value at 3 kHz, the phase modulation reduces to half of the value at ~10 kHz.

Fig. 5a shows the amplitude of the phase modulation as



Fig. 4 a An example of MZI output waveform with modulation frequency of 3 kHz. b Normalized frequency response of the MIR phase modulator.

is

a function of power level of the control beam at the modulation frequency of 3 kHz for the gas pressure of 2 bar. The phase modulation increases nonlinearly with control power level and reaches π rad at the control power of ~280 mW. The minimum HCF length for π rad phase modulation of the MIR beam is estimated to be 7.7 cm. This is because nearly 99% of the control light is absorbed by the pure C_2H_2 at this length, and longer fibers may have little effect on the photo-thermal phase modulation. Furthermore, as control power increases, the gas temperature in the HCF also increases. We believe that the temperature change at high control power is responsible for the nonlinear response^{23,28}. Simulated distribution of temperature inside the HCF using COMSOL Multiphysics indicates that the temperature may change up to 100 K in the core center of the HCF. Here we assume that the temperature changes from 300 to 400 K, the temperaturedependent absorption coefficient is reduced from 0.6 to 0.4 cm^{-1} (33%), the gas density is reduced from 1.04 to 0.78 kg/m^3 (25%), the thermo-optical coefficient is reduced from -1.82×10^{-6} to -1.365×10^{-6} K⁻¹ (25%), the heat capacity is increased from 1699 to 1938 J/(kg·K) (14%), and the heat conductivity is increased from 0.0214 to $0.0333 \text{ W/(m \cdot K)}$ (55%). It should point out that all these factors have negative gain on the phase modulation, which jointly responsible for the nonlinear behavior.

Fig. 5b shows the amplitude of phase modulation at 3 kHz with the control power of 650 mW for different gas pressure inside the AR-HCF. The phase modulation increases approximately linearly with gas pressure. The gas molecular density increases with the pressure level, which leads to increased phase modulation. The phase modulation reaches 2.2π rad for the gas pressure of 3.4 bar.

MIR intensity modulator performance

The MIR all-optical intensity modulation



demonstrated with the gas pressure of 2 bar. We first adjust the two irises to balance the power of the two interference arms as much as possible so that the interference minima are close to zero. Instead of operating at quadrature, here the MZI is stabilized at an interference minima by use of a first harmonic (1*f*)-locking method described in Ref. 29. The piezo-mounted mirror is dithered at 1 kHz and the first harmonic is detected using a lock-in amplifier, which is used as the error signal for the servo-loop with a bandwidth of 1 Hz.

To characterize the modulation/switching feature of the intensity modulator, the control beam is modulated via the AOM with square wave of tunable repetition rate. Fig. 6a shows the measured waveforms of the input control beam and output signal beam with a repetition frequency of 20 Hz and a duty cycle of 50%. The response time of the intensity modulator is estimated following the 10%-90% rule³⁰. Time constants for the rising and falling edges are evaluated to be 1.8 ms and 1.2 ms, respectively, which are limited by the heat dissipation process in the gas-filled AR-HCF. The "on-off" switching capability of the modulator is also evaluated. Without the control beam, the MZI IM output keeps near zero, corresponding to "off" state. With modulated control beam, the IM output changes periodically between zero and the maximum. Here we define the on-off ratio of the IM as the ratio of the maximum value to the minimum value of the IM output, as $10lg(V_{max}/V_{min})$. Fig. 6a shows the measured IM output with a relatively low "off" value under the conditions of control beam power of 82 mW and modulation frequency of 20 Hz. An on-off ratio value of ~25 dB is achieved.

To further investigate the frequency dependence of the intensity modulation, the repetition frequency is varied from 10 Hz to 10 kHz and some of the corresponding signal waveforms are shown in Fig. 6b. As the modulation frequency increases, the rise/fall times are basically unchanged, but the signal waveform gradually changes from a square wave to a sinusoidal wave. The amplitude is found reduced by nearly 80% at 10 kHz, which may be limited by the thermal dynamics in the gas-filled HCF.

NIR phase and intensity modulator performance

The ultra-broadband low-loss transmission of the AR-HCF means that the fabricated modulators could also work well in the NIR band. We test this by using 1.55 μ m fiber laser as the signal beam, which is launched into the system through the WDM, as indicated in the dotted line in Fig. 2. To achieve high coupling efficiency between NIR signal beam and the AR-HCF, the spot parameter of the focused beam is optimized to match the hollow-core diameter of the AR-HCF. The MIR photodetector is replaced by an NIR photodetector.

Fig. 7a, b show, respectively, the phase modulation as functions of control light power and gas pressure. At the control frequency of 3 kHz and AR-HCF gas pressure of 3 bar, a maximum phase modulation of 3.4π rad is obtained at the control power of 650 mW. The phase modulation increases linearly with pressure in the range of 1-3.5 bar for the control light power of 82 mW.

Fig. 8a shows the intensity modulation waveform at the frequency of 100 Hz and control power of 82 mW. The rising time and falling time are 1.8 ms and 1.2 ms, the same as the MIR intensity modulator. The waveforms for increasing modulation frequency from 50 Hz to 10 kHz are shown in Fig. 8b, similar to those of the MIR intensity modulation.



Fig. 6 a waveforms of input control and output signal beam at 20 Hz. **b** Measured signal waveforms with repetition frequency from 10 Hz to 10 kHz.





Discussion

In Table 1, we summarize parts of the performances of the current commercial and state-of-the-art MIR phase/intensity modulators^{13,14,31-33}. Commercially available MIR phase modulator can operate modulation speed of up to 20 GHz, but it can only work at one fixed resonant frequency without the wideband tunable function³¹. In addition, the maximum amplitude of the phase modulation is less than π rad in the MIR band. Furthermore, the damage threshold of the commercial PM is about ~1 W/mm², while the damage threshold of the gas-filled HCF PM can reach up to $>10^6$ W/mm² level, nearly 6 orders of magnitude larger than the commercial one³⁴. Therefore, the wide-band gas-filled HCF MIR modulator is promising for high-power application, which is challenging to achieve with solid state materials.

State-of-the-art modulators based on different mechanisms have been developed^{13,14,32,33}, but most MIR

modulators are electrically driven by external electronics applied directly on the modulation device. Comparing with electrically driven modulators, the gas-filled HCF modulator enable all-optical modulation, within which the signal beam is modulated by the control beam in photonic domain without applying any external electronic or other physics effects. Besides, in this work, a cost-efficient laser in the telecom band is used as the NIR control beam to achieve phase and intensity modulation over an ultra-broad band from NIR to MIR. Due to its own material properties and heat conduction process, the bandwidth and modulation frequency of the gas-filled HCF MIR modulator is currently limited to kHz-level, relatively slower than that of crystal or waveguide-based modulators. Nevertheless, the proposed modulator has almost the widest operating spectral range from NIR to MIR and relatively high intensity modulation on-off ratio. With respect to the effects of nonlinear absorption and

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Ref.	Materials	Effect	Phase modulation (PM)	Modulation bandwidth	On-off ratio (IM)	Wavelength band	Insertionlos
31ª	Crystal	Pockels ^c	<1π	Resonant frequency ^e	NA	1-3 or 3.1-7.4 or 7.5-12 μm ^f	Not stated
32 ^b	Ge-on-Si waveguide	Electro- absorption ^c	0.75π	60 MHz	13 dB	3.8 µm	Not stated
33 ^b	Ge-on-Si waveguide	free-carrier absorption ^d	NA	55 MHz	5.1 dB	2-3 μm	Not stated
13 ^b	Sol waveguide	thermo-optic [°]	NA	21.3 kHz	17.8 dB	3.8 µm	11.8 dB
14 ^b	SiLN waveguide	Pockels	NA	23 kHz	8 dB	3.39 µm	3.3 dB
This work	C_2H_2 in HCF	Photothermal ^d	2.2π	10 kHz	>25 dB	1-4 μm ⁹	3 dB (PM)

 Table 1
 The performance of current commercial and state-of-the-art MIR phase/intensity modulators.

a: the commercial modulator, b: state-of-the-art modulator, c: electrically driven modulator, d: all-optical modulator, e: The user can only select one fixed resonant frequency from 0.1 MHz to 20 GHz, f: The working spectral range is discontinuous, users can only purchase one or more devices in the discrete 1-3 or 3.1-7.4 or 7.5-12 μ m bands, g: The transmission losses (less than 0.5 dB/m) are measured at several wavelengths, and the actual transmission range is broadband, which can reach at least 1-4 μ m. Ge-on-Si: germanium-on-silicon, SoI: Silicon-on-insulator, SiLN: silicon-on-lithium-niobate, NA: Not applicable.

refraction, the former is relatively weak in gas-filled HCF even with light intensity of up to hundreds of MW/cm² level³⁴. For the latter effect, the nonlinear rafractive index (n₂) of gas is in the ~10⁻¹⁹ cm²/W level, which is nearly 7-15 orders lower than that of solid state materials (~10⁻⁴ to 10^{-12} cm²/W level)^{35,36}. Therefore, these two effects in gas-filled HCFs are indeed much weaker than in solid state materials.

It is worth pointing out that the application of the kHzlevel MIR modulator may be limited in the field of communication field, but they may have other potential applications where fast modulation is not required⁶. Actually, this issue can be alleviated by using an HCF with a smaller core diameter so that heat conduct to the environment via the fiber cladding is faster to achieve modulation bandwidth of MHz or higher. Additionally, by using an HCF filled gas with a larger thermo-optic coefficient, and at a higher gas pressure, the control power level required to produce π rad of phase modulation could be reduced. The control power required for π rad phase change in the MIR is larger than that in the NIR, which is expected because the phase change, for the same RI modulation, is inversely proportional to the signal wavelength λ .

In summary, we have demonstrated all-optical phase and intensity modulators from NIR to MIR based on photothermal effect in a C₂H₂-filled AR-HCF. The phase of the signal beam is directly modulated by a NIR control beam, leading to signal phase modulation up to 2.2π rad in the MIR and 3.4π rad in the NIR range. By using a powermatched MZI together with this phase modulator, we have also demonstrated an intensity modulator with on-off ratio of 25 dB. This work brings HCF to the field of MIR modulator devices. The gas-filled HCF may be used as an effective platform to expand the scope of MIR modulators to achieve new photonic functionalities.

Materials and methods

Fabrication of the AR-HCF gas cell.

The AR-HCF gas cell used in the experiment is made of aluminum. The MIR/NIR beam is coupled into the AR-HCF via optical collimator and convex lens as demonstrated in Fig. 2. The AR-HCF is inserted into a ceramic ferrule and the ferrule is embedded in the homemade gas cell. The two ends of the AR-HCF are sealed with CaF₂ windows for light coupling. The AR-HCF is also aligned to the input control and signal beam, and the relative positions are adjusted by use of a 3-axis translation stage under the gas cell to achieve a reasonable coupling efficiency. For gas filling purpose, gas inlet and outlet are equipped on both sides of the cell, and a gap of ~1 mm is kept between the CaF₂ windows and HCF, which allows pure C₂H₂ filling into the hollow-core area.

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Author contributions

W.J. and S.J. proposed the original idea. KZ. built the systems and conducted the experiments. KZ., S.J., F.C., and Y.Z. analyzed the results and prepared the manuscript. S.G., and Y.W. designed and fabricated the AR-HCF. H.L.H. and W.J. supervised the project. The manuscript was discussed and corrected by all authors.

Conflict of interest

The authors declare that they have no conflict of interest.

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