Photoacoustic Spectroscopy of Gas Filled Hollow Core Fiber

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Abstract: Photoacoustic spectroscopy is demonstrated with gas filled microstructured hollow core optical fibers. This technique may be used for high sensitivity gas sensing, non-invasive fiber characterization, and fiber-optic phase modulation devices © 2022 The Authors.

1. Introduction

Photoacoustic spectroscopy (PAS) is a powerful technique for gas detection by measuring the acoustic wave generated due to optical absorption. Traditionally, an open-path PAS cell is used for light absorption and acoustic wave generation, and a microphone for acoustic detection. By use of an acoustic resonator [1], an optical resonating cavity [2], or a specially designed ultra-sensitive microphone (e.g., interferometric cantilever [3]), high sensitivity gas detection has been demonstrated.

Here, we report PAS of a gas-filled hollow-core fiber (HCF) [4]. As shown in Fig.1, gas absorption of a modulated pump beam in the hollow core generates acoustic wave, which is resonantly amplified in the silica microstructures and modulates the phase of a probe beam propagating in the fiber. The HCF acts as a compact gas cell for pump absorption and acoustic generation, an acoustic resonator, and an acoustic detector. The photoacoustic wave modulates the phase of a probe beam, which is detected by an optical interferometer. The optical spectrum of the phase modulation at an acoustic resonance offers a means of photoacoustic gas spectroscopy with enhanced sensitivity. And the acoustic resonances measured by the PAS can be used to characterize the inhomogeneity along the fiber.



Fig.1 Basic setup of PAS of a gas filled HCF.

2. Principle

Fig. 2(a) shows an anti-resonant HCF (AR-HCF) used in our experiment. It consists of an air-core and seven suspended silica capillaries of slightly different sizes. The structure of a gas-filled AR-HCF is a vibroacoustic system that supports acoustic modes in air (air mode) and silica capillaries (capillary mode). They form hybrid acoustic modes.



Fig. 2. (a) SEM image of AR-HCF. (b) capillary mode. (c) air mode. (d) optical modes (LP₀₁, LP_{11a}, and LP_{11b}) [4].

Finite-element-method (FEM) analysis based on thermo-viscous acoustic model shows the gas-filled AR-HCF supports several coupled acoustics modes in the megahertz range. As an example, Fig. 2(b) shows a capillary mode (the first-order wine-glass-like mode) with an eigenfrequency of 4.50 + 0.018i MHz, where the imaginary

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part stands for the loss of the mode. An air mode (the first-order radial-like mode) with an eigenfrequency of 5.15 + 0.70i MHz is shown in Fig. 2(c). Compared with the capillary mode, the air mode is relatively lossy.

The acoustic modes are excited by the photoacoustic effect due to gas absorption of the modulated pump beam. The phase of the probe is modulated by the acousto-optical interaction over the length of the fiber. Figure 2(d) shows the optical modes (LP₀₁ and LP₁₁) supported by the AR-HCF. The fundamental mode (LP₀₁) and the high-order mode (LP₁₁) have different overlaps with the acoustic modes, and hence the phase of the two probe optical modes experience different modulation along the fiber. By detecting the phase modulation of either of the modes, the acoustic modes and/or gas absorption can be probed. Here, we use an in-fiber modal interferometer to detect the differential phase modulation between the two probe modes, which has proven to be insensitive to external disturbance due to common-path noise cancellation [5] but quite sensitive to the acoustic modes due the different overlap of the probe optical modes and the acoustic modes. The differential phase modulation may be expressed in the form of [4]

$$\Delta\phi(\Omega,\lambda_P) = \xi(\Omega)\alpha(\lambda_P)CLP_P \tag{1}$$

where $\alpha(\lambda_P)$ the absorption coefficient at the pump wavelength λ_P , *C* the gas concentration, *L* the length of the fiber, P_P the power of pump, and $\xi(\Omega)$ is a normalized phase modulation coefficient that describes the frequency response of the fiber at the pump modulation frequency Ω . $\xi(\Omega)$ can be enhanced significantly near the acoustic resonances. By measuring $\alpha(\lambda_P)C$ and $\xi(\Omega)$, the proposed method can be used for photoacoustic gas spectroscopy and spectroscopy of fiber microstructure, respectively.

3. Experimental results

Experiment was conducted with a 30-cm-long AR-HCF (fiber A). The AR-HCF is firstly filled with 10 mbar pure C_2H_2 . The pump laser is centered near the P(13) absorption line of the $v_1 + v_3$ band acetylene at 1532.83 nm. A lock-in amplifier is used to detect the harmonic signal at the modulation frequency $f = \Omega/2\pi$. Figure 3(a) shows the measured phase modulation in the frequency range from 25 kHz to 7 MHz when the pump is tuned to the absorption line (blue trace) and detuned from the absorption line (purple trace). Seven strong resonances were observed around 5 MHz, corresponding to the seven capillaries of the AR-HCF. We also measured the gas absorption spectrum by fixing the pump modulation frequency at an acoustic resonance near 4.51 MHz while sweeping the pump wavelength across the gas absorption line. As shown in Fig. 3(b), a Doppler limited lineshape is observed, and an isotope of C_2H_2 is resolvable.



Fig. 3. PAS spectra of a 30-cm-long AR-HCF filled with 10 mbar pure C₂H₂ [4].

We also conducted an experiment with another piece of 30-cm-long AR-HCF (labeled as fiber B), a different segment of the same AR-HCF. The experiment result is shown in Figs. 3(c) and (d). The two HCFs show similar acoustic resonances. However, due to the scaling of the fiber structure during the drawing process, the resonances for fiber B are upshifted by about 8%. The enlarged pictures of the gray shaded region of PAS spectra near 4.55 MHz and 4.95 MHz are shown in Fig. 3(d). The quality factor (Q-factor) of the acoustic modes of fiber A and fiber B are determined to be 480 and 256, respectively. FEM simulation shows the Q-factor limited by gas damping is about 940. Hence, the Q-factors of the two AR-HCF are limited by the inhomogeneity of the silica

capillary along the fiber. From the multi-peak spectra of the two fibers, we can calculate the longitudinal inhomogeneity by FEM analysis. For fiber A, the frequency difference between the two peaks is about 6 kHz, corresponding to the inhomogeneity of 1.6%/m in the wall thickness or 2.4%/m in the capillary diameter. This value is close to the inhomogeneity estimated by the relative change of the resonant frequency (about 1.7%/m). For fiber B, the inhomogeneity is estimated to be 2.3%/m in the wall thickness or 3.6%/m in the capillary diameter.

Gas detection was performed with fiber A. The AR-HCF was filled with 106 ppm C_2H_2 at a pressure of 1 bar. At this pressure, the Q-factor of the resonance near 4.51 MHz is ~120. The modulation frequency of pump is fixed at 4.51 MHz and the wavelength of pump is slowly swept across the absorption line of C_2H_2 near 1532.83 nm. Fig. 4(a) shows the measured absorption spectra on-resonance at 4.51 MHz (blue curve) and off-resonance at 4.3 MHz (red curve). Compared with the non-resonant background, the photoacoustic signal at 4.51 MHz is significantly enhanced. As shown in Fig. 4(b), the measured signal is proportional to the pump power, but the noise is independent of pump power. The maximum phase modulation is estimated to be 214 μ rad with a 162mW pump power, corresponding to a normalized phase modulation coefficient of $\xi_{max} = 0.39 rad/W$. Allan analysis based on the noise data obtained with the pump away from the absorption line gives a noise-equivalent concentration of ~8 ppb with an integration time of 100 s.



Fig. 4. Results of gas detection experiment with 106 ppm C₂H₂ balanced with nitrogen [4].

3. Discussion

In summary, we demonstrated PAS of gas-filled AR-HCFs. By employing a wine-glass-like acoustic mode of the silica capillary, we demonstrated a PAS for gas detection with a noise-equivalent concentration of 8 ppb C_2H_2 . The acoustic resonances detected by PAS can be used to characterize fiber inhomogeneity. Experiments with two segments of the same AR-HCF reveal longitudinal inhomogeneity of a few percent per meter along the fiber.

The sensitivity of PAS demonstrated here can be further improved by improving the phase sensitivity to the deflection of fiber microstructure. For the AR-HCF we used, the phases of the high-order optical modes are more sensitive to the capillary mode. The maximum displacement of the capillary mode is estimated to be $\delta = 2.9$ pm, corresponding to a sensitivity to the deflection of $|\partial n_{11b}/\partial \delta = 41 \ m^{-1}|$ for the most sensitive LP_{11b} mode. The sensitivity can be enhanced by optimizing the waveguide structure. For example, a sensitivity of $|\partial n_{11b}/\partial \delta = 64 \times 10^3 \ m^{-1}|$ has been demonstrated on a dual-nanoweb fiber [6]. A potential enhancement of the PAS signal by a few orders of magnitude is possible under atmospheric condition.

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