Supplemental Document

Photoacoustic Brillouin spectroscopy of gas-filled anti-resonant hollow-core optical fibers: supplement

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1. GEOMETRY OF AR-HCF MODEL BUILT FROM SEM

The geometry model of anti-resonant hollow-core fiber (AR-HCF) used for simulation is precisely built from the scanning electron microscopy (SEM) of the fiber. SEM image of fiber A and its geometry used for simulation are shown in Figs. S1(a) and (b), respectively. The simulated electric field \( \psi \) is shown in Fig. S1(c).

Fig. S1. (a) SEM image of fiber A. The white bar is 20 \( \mu m \). (b) Geometry of fiber A used for simulation. The capillaries of fiber A are labeled from 1 to 7. (c) Calculated electric field \( \psi_i (i = 01, 11a, 11b) \) with nitrogen of 1 atm at 1532.83 nm and the corresponding mode inices.

The diameter of the whole hollow region is 60.5 \( \mu m \) and its center is set at (0, 0). The inner diameter, thickness and center of capillaries estimated from SEM are tabulated in Table. S1.

| Label | Inner diameter (\( \mu m \)) | Thickness (nm) | Center of the capillary (\( \mu m, \mu m \)) |
|-------|-----------------------------|---------------|-----------------------------------------------|
| 1     | 15.10                       | 406           | (-6.5,-22)                                   |
| 2     | 14.26                       | 439           | (-22.4,5.6)                                  |
| 3     | 13.78                       | 459           | (-18.2,14.9)                                 |
| 4     | 13.89                       | 443           | (0.4,23.4)                                   |
| 5     | 13.70                       | 395           | (18.3,15.3)                                  |
| 6     | 13.90                       | 428           | (23.3,-2.5)                                  |
| 7     | 14.28                       | 413           | (12.5,-19.7)                                 |
2. PUMP AND HEAT SOURCE IN AR-HCF

A. Absorption coefficient of acetylene

We focus on the P(13) line in the $\nu_1 + \nu_3$ band of acetylene (C$_2$H$_2$) at 1532.83 nm. Foreign gas broadening affects the absorption coefficient ($\alpha$) of C$_2$H$_2$. For simplicity, we assume nitrogen has the same foreign broadening coefficient with that of air. The absorption coefficients of C$_2$H$_2$ versus gas pressure are shown in Fig. S2. They are calculated based on the HITRAN database [1]. In the Doppler regime (i.e. pressure less than 1 kPa), the wall collisional broadening in AR-HCF estimated with hard sphere model is about 7 MHz [2, 3], which is much smaller than the Doppler linewidth and is ignored in the simulation.

![Fig. S2](image)

**Fig. S2.** The absorption coefficient at 1532.83 nm at different gas pressures for pure C$_2$H$_2$ and trace C$_2$H$_2$ in nitrogen. The absorption coefficient of nitrogen balanced C$_2$H$_2$ is normalized by its concentration. The increase of absorption coefficient for pressure higher than 1 bar is due to the neighbor absorption lines.

B. Thermal relaxation time of acetylene

Thermal relaxation time ($\tau$) characterizes the speed of heat generation after optical absorption. It determines the efficiency of generating a harmonic heat source in the MHz regime. For C$_2$H$_2$, it is determined by the V-R, T relaxation process and is inversely proportional to gas pressure $p$ [4]. We measure the thermal relaxation processes in AR-HCF with a 10-ns optical pulse and the result is shown in Fig. S3. The result yields a pressure-dependent thermal relaxation time of:

$$p\tau = 74 \text{ ns} \cdot \text{atm}(N_2 \text{ balanced } C_2H_2) \quad (S1)$$

![Fig. S3](image)

**Fig. S3.** The transient response of thermal relaxation and thermal conduction in AR-HCF. The fiber is filled with 106 ppm C$_2$H$_2$ balanced with nitrogen.
C. Pump light and heat source

AR-HCF mainly supports two sets of LP-like optical modes as shown in Fig. S1(c). The intensity of the pump is modulated by an acoustic optical modulator. The optical intensity of pump is

\[ I_p = I_0 + \bar{I} \]  
\[ I_0 = P_p [\Psi_1 + \Psi_2 \cos(k_p n_r z)] \]  
\[ I \approx P_p [\Psi_1 + \Psi_2 \cos(k_p n_r z)] \exp(-j\Omega t) \]  

where \( \Psi_1 = \eta \psi_{10}^2 + (1 - \eta) \psi_{11}^2 \), \( \Psi_2 = 2\sqrt{\eta (1 - \eta)} \psi_{101} \psi_{11} \), and \( \psi_1 \) are the power normalized electric field of \( LP_{10} \) and \( LP_{11} \)-like modes, \( \eta \) the fractional power in \( LP_{10} \), \( P_p \) the pump power, \( z \) the direction of light propagation, \( k_p \) the angular wavevector of pump light, \( n_r = n_{10} - n_{11} \) the differential mode index between \( LP_{10} \) and \( LP_{11} \) and \( \Omega = 2\pi f \) is the angular frequency of pump modulation. For AR-HCFs, more than 99.9\% power of light is in the gas (\( n_{gas} \approx 1 \)) and hence it is a good approximation to normalize that \( \int dS \bar{\rho}^2 \approx 1 \) over the fiber cross-section. For modulation frequency of a few MHz and fiber length of meters or shorter, the phase term of optical intensity \( g_L \approx 2\pi f L/c \ll \pi \) (where \( L \) is the length of fiber) and could be ignored (the effective mode indices are all very close to unity). For simplicity, the heat source is estimated with a two-level model and is expressed by

\[ Q = Q_0 + \tilde{Q} \]  
\[ Q_0 = \frac{a C L_0}{\sqrt{\Omega^2 \tau^2 + 1}} \]  
\[ \tilde{Q} = \frac{a C I}{\sqrt{\Omega^2 \tau^2 + 1}} \exp(j\phi) \]  

where \( Q_0 \) is the DC component of the heat source, \( \tilde{Q} \) the harmonic heat source, \( C \) the concentration of \( C_2H_2 \) and \( \phi = \tan^{-1}(\Omega \tau) \) is the phase angle.

3. THERMO-VISCOUS ACOUSTICS IN AR-HCF

For simulation of acoustic problems in small dimension, viscous and thermal dissipation at the boundary play an important role and a thermo-viscous acoustic model is often used [5]. Meanwhile, we need to consider the coupling between the thermo-viscous acoustics and the silica capillaries [6]. The thermo-viscous model and the solid mechanics model are used for simulation in COMSOL Multiphysics. The parameters used in the simulation is listed in Table. S2 [7–10]. The density of gas is assumed to be linear dependent on gas pressure. The thermo-viscous acoustic model is coupled with the solid mechanics model with an isothermal boundary condition. For low concentration of \( C_2H_2 \) in nitrogen, parameters of \( N_2 \) is used for simplicity.

The steady state is calculated by assuming \( Q = Q_0 \). For simplicity, we ignore the deformation of the fiber geometry at steady state and only consider the steady state temperature field \( T_0 \) and pressure field \( p_0 \). Then we calculate the harmonic perturbation in the frequency domain. The harmonic perturbation is accurate only when the harmonic perturbations of pressure, temperature and density are much smaller than their steady state values. Since the variation of heat source along z-axis is much smaller than in the fibre cross-section, hence the simulation is performed for simplicity with a 2-D model. We have

\[ \rho = [\rho_1 + \rho_2 \cos(k_p n_r z)] \exp(-j\Omega t + \phi) \]  
\[ w = [w_1 + w_2 \cos(k_p n_r z)] \exp(-j\Omega t + \phi) \]  

where \( \rho_1 \) and \( w_1 \) are the harmonic solutions when only the first term of \( I \) on the right hand side (RHS) of Eq. S4 is applied, \( \rho_2 \) and \( w_2 \) are the harmonic solution when only the last term of \( I \) on the RHS of Eq. S4 is applied.

As demonstrated in Fig. S4, the modal interference gives a periodic optical intensity distribution \( I_p = I_0 + \bar{I} e^{-j\Omega t} \) and hence the heat source distribution \( Q = Q_0 + \bar{Q} e^{-j\Omega t} \) as shown in Fig. S4(a) along fiber with a period of \( d = \lambda_p / n_r \approx 1.6mm \). The periodic heat source results in a periodic distribution of acoustic field, which is described by the density change \( \rho = \rho_0 + \bar{\rho} e^{-j\Omega t} \), and the displacement of capillary \( w = w_0 + \bar{w} e^{-j\Omega t} \). In our experiment, \( L/d \gg 1 \) and hence the total phase modulation is equivalently induced by the equivalent acoustic fields \( \bar{\rho} = \rho_1 + \bar{\rho} \) and \( \bar{w} = w_1 \). The cosine terms in Eq. S8 and Eq. S9 are averaged out. The acoustic fields \( \tilde{\rho} \) and \( \tilde{w} \) are induced by an equivalent harmonic pump \( \bar{Q} \). The equivalent fields (\( \tilde{\rho}, \tilde{w} \) and etc.) are equal to the fields at the quarter period as demonstrated in Fig. S4.
Table S2. Parameters used for acoustic simulation

| Symbol | Description                     | Value at T₀ |
|--------|---------------------------------|-------------|
| N₂     | Dynamic viscosity               | 1.8 × 10⁻⁵ Pa · s | 1.0 × 10⁻⁵ Pa · s |
| C₂H₂   | Dynamic viscosity               | 1.4 × 10⁻⁵ Pa · s | 7.0 × 10⁻⁴ Pa · s |
| µ      | Bulk viscosity                  | 1.0 × 10³ J/(kg · K) | 2.2 × 10³ J/(kg · K) |
| c_p    | Isobaric specific heat capacity | 0.026 W/(m · K) | 0.024 W/(m · K) |
| κ      | Thermal conductivity            | 348 m/s     | 343 m/s     |
| ν      | Speed of sound                  | 1.165 g/L   | 1.092 g/L   |
| ρ₀     | Gas density and 1 atm           | 1 + 2.8 × 10⁻⁴ | 1 + 5.5 × 10⁻⁴ |
| γₚ     | Ratio of specific heat capacity | 1.4         |             |
| K_T    | Isothermal compressibility      | γₚ/(ρ₀µ²)   |             |
| T₀     | Ambient temperature             | 293.15 K    |             |
| αₚ     | Isobaric thermal expansion coefficient | 1/T₀          |             |
| ρₛ     | Density of fused silica         | 2203 kg/m³  |             |
| E      | Young’s modulus of fused silica | 73.1 GPa    |             |
| nₛ     | Refractive index of fused silica | 1.444       |             |

Fig. S4. The modal interference induced periodic distribution of (a) harmonic heat source $\tilde{Q}$, (b) density change $\tilde{ρ}$, and (c) displacement of capillary $\tilde{w}$. The parameters are $a = 1.06$ cm⁻¹, $C = 106$ ppm and $Pₚ = 162$ mW. The displacement in the figure is enlarged by $2 \times 10⁶$ times for clarity. The slice long the fiber is plotted along the dash line of the fiber cross-section.
4. PROBE THE ACOUSTIC MODES WITH AN OPTICAL WAVE

A. Phase modulation of probe beam

The probe beam in the AR-HCF experiences phase modulation (or modulation of mode index \( n_m \)) due to shifting boundary (SB), gas density change (GD) and photoelasticity (PE):

\[
\frac{dn_m}{dx} = \frac{dn_m}{dx}_{SB} + \frac{dn_m}{dx}_{GD} + \frac{dn_m}{dx}_{PE}
\]  

(S10)

where \( x \) is an infinitesimal perturbation (i.e. a normalized equivalent harmonic heat source \( \dot{Q}/\alpha CP \)) and subscript \( m \) refers to the label of optical modes \((m = 01, 11a, 11b)\). Since we only care about the harmonic phase modulation near the steady state, we only consider the phase modulation due to harmonic perturbations. Considering the pump depletion due to optical absorption and ignoring the fiber loss, the phase modulation is calculated by

\[
P(z) = P_0 \exp(-\alpha CL)
\]

(S11)

\[
\Delta \phi_m = \frac{\alpha C \omega_L}{c} \int_0^L dz P(z) \frac{dn_m}{dx}
\]

(S12)

where \( L \) is the length of AR-HCF and \( dn_m/dx \) is independent of \( z \) when considering a normalized equivalent heat source \( \dot{Q}/\alpha CP \). Hence from Eq. S11 and S12 we have,

\[
\Delta \phi_m \approx \frac{P_0 \omega_L}{c} \alpha CL \frac{dn_m}{dx}
\]

(S13)

\[
L_{eff} = \frac{1 - e^{-\alpha CL}}{\alpha C}
\]

(S14)

where \( L_{eff} \) is the effective absorption length and it approximates the length of fiber \( L \) when \( \alpha CL \ll 1 \). The modulation of refractive index is related with modulation of eigenfrequencies of optical modes by [11]

\[
\frac{dn_m}{dx} = -\frac{n_m^4}{\omega_L^2} \frac{dn}{dx}
\]

(S15)

where \( n_m^k = n_i + dn_i/dx \) is the group mode index and \( \omega_L \) is the angular frequency of the probe. The shifting boundary induced perturbation is calculated as [12]

\[
\left. \frac{dn_m}{dx} \right|_{SB} = \frac{n_m^4}{2} \int dS (w \cdot \hat{n}) [\Delta \epsilon_{12} \hat{n} \times E_m]^2 - \Delta (\epsilon_{12}^2) [\hat{n} \cdot D_m]^2]
\]

(S16)

where \( \hat{n} \) is the normal vector on the interface from silica to gas, \( E_i \) the electric field, \( D_m = eE_m \) the electric displacement field, \( \Delta \epsilon_{12} = \epsilon_1 - \epsilon_2 \) and \( \Delta (\epsilon_{12}^2) = \epsilon_1^{-1} - \epsilon_2^{-1} \) are the differential permittivity between silica (\( \epsilon_1 \)) and gas (\( \epsilon_2 \)). The density change and photoelasticity induced perturbations are calculated by the standard perturbation theory [13, 14]

\[
\left. \frac{dn_m}{dx} \right|_{GD} = \frac{n_m^4 \gamma_c}{2 \rho_0} \frac{\int dS |E_m|^2 \rho}{\int dS |E_m|^2 \epsilon}
\]

(S17)

\[
\left. \frac{dn_m}{dx} \right|_{PE} = \frac{n_m^4 n_{silica}^4 \varepsilon_0}{2} \frac{\int dS \left( \sum 2Re(E_{i,m}^* E_i) p_{44} S_{ij} + \sum |E_{i,m}|^2 (p_{11} S_{ij} + p_{12} (S_{ij} + S_{kk})) \right)}{\int dS |E_m|^2 \epsilon}
\]

(S18)

where the electrostrictive constant \( \gamma_c = 2(\eta_{gas} - 1) = A \rho_0 \) for dilute gas and \( A \) is a constant [13]. \( S_{ij} \) is the strain tensor and for fused silica, \( p_{11} = 0.121, p_{12} = 0.27 \) and \( p_{44} = (p_{11} - p_{12})/2 \) [14]. The phase modulation of modal interferometer (MI) is

\[
\Delta \phi_{MI} = \Delta \phi_{01} - \Delta \phi_{11a,b}
\]

(S19)

For example, Fig. S5 shows the simulation result with 106 ppm \( C_2 H_2 \) balanced with nitrogen at a pressure of 1 bar. As shown in Fig. S5(a), for optical modes in the gas, the contribution of phase modulation from photoelastic effect could be ignored. The contributions from gas density change and shifting boundary are comparable. Different optical modes also experience different phase modulation as we shown in Fig. S5(b). However, the two polarizations of an optical mode shows little difference as we found in the simulation.
**Fig. S5.** The calculated phase modulation with $P_p = 162 \text{ mW}$, $C = 106 \text{ ppm}$ and $L = 30 \text{ cm}$. (a) The phase modulation due to different mechanisms for $LP_{01}$ mode. (b) The phase modulation of different optical modes.

**B. Calibration of the phase modulation**

The light from AR-HCF is coupled into a single-mode-fiber (SMF) and forms a modal interferometer. The phase modulation of probe is calibrated with the interferogram measured by detuning the wavelength of probe (Fig. S6). The lock-in amplifier measures the root-mean-square (RMS) voltage of the small harmonic signal ($V_{RMS}$). Hence, the experimental phase modulation could be calibrated as

$$|\Delta \phi_{exp}| = \frac{1.4V_{RMS}}{V_0}$$

**Fig. S6.** The measured interferogram of the modal interferometer. The wavelength of probe is centered at the quadrature point (red dot). In the experiments, $V_0 = 0.8 \text{ V}$ and $V_{DC} = 1.8 \text{ V}$ for experiment with $106 \text{ ppm } C_2H_2$.

**C. Raman gain of SMF pigtails**

The harmonic pump modulates the Raman gain on the probe. The harmonic Raman gain is detected by the lock-in amplifier and then gives an equivalent background ‘phase modulation’. The Raman gain with a frequency difference $\Delta \nu = \nu_p - \nu_L = 72 \text{ cm}^{-1}$ is about 0.14 of its peak value [15]

$$g_R(\Delta \nu) = 0.14g_{R,\text{peak}} \approx 7.8 \times 10^{-15} \text{ m/W}$$

In the experiment setup, the harmonic pump power is about $P_p = 162 \text{ mW}$. The length of SMF pigtails is about $l_{SMF} = 25 \text{ cm}$ and the effective cross-section of SMF is about $A_{eff} = 76 \mu m^2$. 
The Raman gain on probe is about
\[ G = g_R (\Delta \nu) P_P \frac{P_P}{A_{eff}} L_{SMF} = 4.2 \times 10^{-6} \] (S22)

The Raman gain induced equivalent background ‘phase modulation’ is
\[ |\Delta \phi_R| = \frac{1.4 G V_{DC}}{V_0} \approx 13 \mu rad \] (S23)

Since the Raman induced background \( \Delta \phi_R \) is incoherent with \( \Delta \phi_{MI} \), the experimental phase modulation \( |\Delta \phi_{expr}| \) is compared with the total phase modulation
\[ |\Delta \phi_{tot}| = |\Delta \phi_{MI}| + |\Delta \phi_R| \] (S24)

5. PRESSURE DEPENDENCE OF PABS OF AR-HCF

As shown in Fig. S7, we measure the PABS spectrum of two different AR-HCFs with a gas pressure from 1 bar to 11 bar. The fiber is filled with 106 ppm \( C_2H_2 \) balanced with nitrogen. The phase modulation due to the radial air mode is approximately proportional to the gas pressure. The shift of the resonances is due to the coupling between the capillary mode and the air mode. Because of the different LP-mode of probe beam, the PABS spectrum at a pressure of 1 bar is different from Fig. 4(a) in the main text.

![Fig. S7. The pressure dependence of the PABS spectrum for two different fiber samples. The gas pressure is increased from 1 bar to 11 bar. The SEM images of the two fibers are shown in Figs. 6(a) and (b) in the main text, respectively. The curves are offset for clarity.](image)

REFERENCES
1. L. S. Rothman, I. E. Gordon, A. Barbe, D. C. Benner, P. F. Bernath, M. Birk, V. Boudon, L. R. Brown, A. Campargue, J.-P. Champion et al., “The hitran 2008 molecular spectroscopic database,” J. Quant. Spectrosc. Radiat. Transf. 110, 533–572 (2009).
2. F. Couny, O. Carraz, and F. Benabid, “Control of transient regime of stimulated raman scattering using hollow-core pcf,” JOSA B 26, 1209–1215 (2009).
3. J. O. Hirschfelder, C. F. Curtiss, R. B. Bird, and M. G. Mayer, Molecular theory of gases and liquids, vol. 165 (Wiley New York, 1964).
4. S. E. Bialkowski, Photothermal spectroscopy methods for chemical analysis, vol. 177 (John Wiley & Sons, 1996).
5. D. T. Blackstock, “Fundamentals of physical acoustics,” (2001).
6. E. H. Dowell, G. Gorman Iii, and D. Smith, “Acoustoelasticity: General theory, acoustic natural modes and forced response to sinusoidal excitation, including comparisons with experiment,” J. Sound vibration 52, 519–542 (1977).
7. E. W. Lemmon and R. T. Jacobsen, “Viscosity and thermal conductivity equations for nitrogen, oxygen, argon, and air,” Int. journal thermophysics 25, 21–69 (2004).
8. A. S. Dukhin and P. J. Goetz, “Bulk viscosity and compressibility measurement using acoustic spectroscopy,” The J. chemical physics 130, 124519 (2009).
9. Z. Gu and W. Ubachs, “Temperature-dependent bulk viscosity of nitrogen gas determined from spontaneous rayleigh–brillouin scattering,” Opt. letters 38, 1110–1112 (2013).
10. M. S. Cramer, “Numerical estimates for the bulk viscosity of ideal gases,” Phys. fluids 24, 066102 (2012).
11. V. Laude, J. M. Escalante, and A. Martínez, “Effect of loss on the dispersion relation of photonic and phononic crystals,” Phys. Rev. B 88, 224302 (2013).
12. S. G. Johnson, M. Ibanescu, M. Skorobogatiy, O. Weisberg, J. Joannopoulos, and Y. Fink, “Perturbation theory for maxwell’s equations with shifting material boundaries,” Phys. review E 65, 066611 (2002).
13. R. W. Boyd, Nonlinear optics (Academic press, 2020).
14. D. Royer and E. Dieulesaint, Elastic waves in solids II: generation, acousto-optic interaction, applications (Springer Science & Business Media, 1999).
15. R. H. Stolen, J. P. Gordon, W. Tomlinson, and H. A. Haus, “Raman response function of silica-core fibers,” JOSA B 6, 1159–1166 (1989).