Supplementary Material

Single-photon laser methane detection methodology and initial validation

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Supplementary Note 1

Theoretical detection model

The fundamental M sequence, along with the cross-correlation of an N-bit PRBS, where $a[i] \in [0, 1]$, and its bipolar sequence can be described by $a'[i] = 2a[i] - 1 \in \{-1, 1\}$. The M-sequence and continuous ramp sweep modulation signals are superimposed on the emitted laser. Then, the emitted laser signal can be written as

$$P_l(i) = P * a[i],\ i \in [1:N] \qquad \text{(S1)}$$

where $P_l(i)$ represents the emission power at the time bin, and i denotes the time bin corresponding to the wavelength sequence. P is laser output power after first modulated by the ramp sweeping. The emitted signals with an average power $P(\lambda_i)$ at different wavelengths are coded with the M sequence. The received signals, dominated by the target range, encounter a time delay τ and are weakened by the aerosol and gas absorption. The received echo photon counts per bin $n_{\lambda i}$ are given by

$$n_{\lambda_i} = P_l(\lambda_i)\eta_l\eta_r\eta_q T_c \rho \frac{\lambda_i}{hc}\frac{A_r}{\pi R^2}\exp(-2OD(\lambda_i)) \qquad \text{(S2)}$$

where $P_l(\lambda_i)$ is the initial laser power emitted at various wavelengths, η_l and η_r are the emission and receiving optical efficiency, respectively, η_q is the quantum efficiency, T_C is the bit time (chip time) of M-sequence, ρ represents the reflectivity of the target (approximate Lambertian apparatus), λ_i denotes the sweeping wavelengths, h is the Planck's constant, c is the speed of light, A_r is the receiving optical unit area, R is the detection distance, and $OD(\lambda_i)$ denotes the total column optical depth in different wavelengths. It can be written as

$$OD(\lambda_i) = \int_0^R [\alpha_a(\lambda_i) + \sigma_{\lambda_i} N_{CH_4}]\, dr \qquad \text{(S3)}$$

where $\alpha_a(\lambda_i)$ is the aerosol extinction coefficient, $\sigma_{\lambda i}$ is the CH_4 absorption cross section at a given wavelength, and N_{CH4} is the number of CH_4 molecules present along the laser integration path.

Supplementary Note 2

Ranging and methane concentration simulation

The results of cross-correlation between the received photon signal $n[i]$ and the M sequence in a bipolar mode for different wavelengths can be written as

$$C_i[\tau/T_c] = n_s \otimes a'[i] = \sum_{k=1}^{N-1} n_s(k) \bullet a'[N+k] \qquad \text{(S4)}$$

The position corresponding to the maximum cross-correlation coefficient can be expressed as $k = \tau/T_c$, where τ is the signal flight time. The laser detection distance can be calculated as

$$R = c * k * T_c / 2 \qquad \text{(S5)}$$

The CH_4 concentrations can be calculated as follows:

Step 1: Based on the cross-correlation results of the received signal and bipolar sequence, determine the flight time τ and the corresponding signal shift bin number k. In Fig. 3, the echo signals at the absorption wavelength have been reduced. The echo signal shift to the wavelength position corresponding to the emitted signal and the recovered signal shift can be calculated as follows

$$n_s\_re(i) = [n_s(k+1:end), n_s(1:k)] \qquad \text{(S6)}$$

where n_s_re is the shifted signal and λ_i is the original emission wavelength sequence with one-to-one correspondence between the two signals.

Step 2: Extract the original signal for a time bin τ_i and wavelength λ_i. For the 10 ns sampling interval, the signal $n_e(\lambda_i)$ can be extracted at the corresponding wavelength.

Step 3: Interpolate the wavelength λ_i sequence and extracted signal $n_e(\lambda_i)$. The interpolated wavelengths λ_{interp} and the signal $n_{interp}(\lambda_i)$ are then baseline-fitted and the baseline-fitted signal is denoted as n_{base}.

Step 4: Divide the data n_{interp} of different wavelengths interpolated from the baseline data n_{base} to obtain the transmittance data $T_{signal} = n_{interp} / n_{base}$.

Step 5: Fit λ_{interp} and T_{signal} to the Lorentz curve drawn with the wavelength λ_{interp} on the x axis and the transmittance T_{signal} on the y axis.

Step 6: Establish the lookup table: Using the theoretical gas-absorption model, the theoretical CH_4 absorption curve is obtained, and the data search matrix of the wavelength, transmittance, and CH_4 concentration is established.

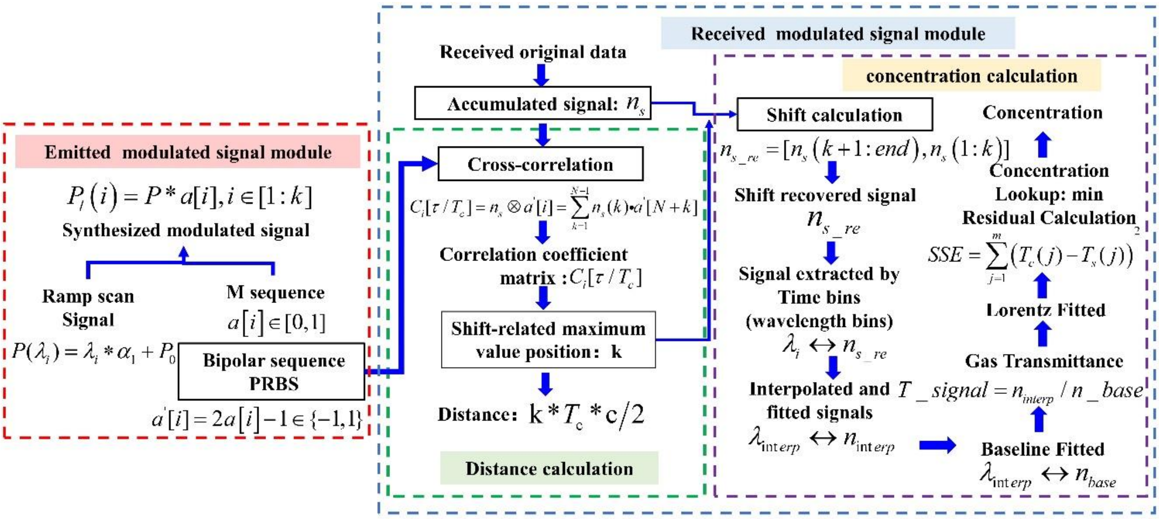

Figure. S1. Inversion algorithm flow for ranging and methane concentration