# All-fiber IPDA lidar for CH<sub>4</sub> leakage monitoring using InGaAs/InP single-photon detector

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**Abstract:** An integrated path differential absorption (IPDA) lidar for CH<sub>4</sub> leakage monitoring is proposed and demonstrated. In the simplified all-fiber optical layout, a homemade InGaAs/InP single-photon detector (SPD) using multi-channel technique with multi-mode fiber coupling is used to increase the maximum count rate and coupling efficiency. The system is calibrated in intensity and frequency domains. Firstly, the fluctuation of the laser power is compensated. Secondly, the dead time, afterpulsing probability and dark counts of the SPD are corrected. A mean relative difference of 0.84% between SPD and PIN photodetector is achieved. Thirdly, non-linear frequency scanning of the laser is measured by homodyne detection and analyzed in joint time-frequency domain. In the symmetry-calibration process, the absorbance spectra of up and down scanning are compared. Maximum difference less than 1% with mean difference of 0.33% is achieved within a span of 4 GHz around the center of absorbance spectrum. Finally, a demonstration experiment over ten days is carried out to analyze the accuracy and stability of the system. A mean deviation of 0.03% with standard deviation of 0.46% is verified at a distance of 12 m and a time resolution of 1 s. By attenuating the laser power from 2 mW to 0.02 mW, the performance of the system is degraded to a mean deviation of 1.32% with standard deviation of 4.33%.

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# 1. Introduction

Global warming is mainly due to greenhouse gas (GHG) emissions from human activities, which has led to numerous extreme weathers across the globe [1–3]. Carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) are the two most important GHGs, contributing more than 63% and 19% of global radiative forcing, respectively [4]. The monitoring of GHGs is extremely important for GHGs emission reduction and climate change mitigation [5]. In the past decades, numerous device development projects have been proposed for efficiently monitoring GHG emissions, such as CO<sub>2</sub> and CH<sub>4</sub> atmospheric remote monitoring-Flugzeug (CHARM-F) and CH<sub>4</sub> remote sensing Lidar mission (MERLIN) [6,7]. Different from the passive remote sensing methods based on ambient light, the above programs adopt active light source that can continue to work at night and is less susceptible to environment interference [8–12]. Different absorption lidar (DIAL) and integrated path differential absorption (IPDA) lidar are widely used for remote sensing of atmospheric gases in open paths [13–16]. Typically, these systems operating at two nearby wavelengths, with one near the center of the line of target gas and the other at the margin with negligible absorption [17]. Multi-frequency DIAL and IPDA lidar can cover one or even more absorption

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lines allowing the simultaneous detection of multiple atmospheric molecules [18–20]. In our previous work, a multi-frequency DIAL has been demonstrated to simultaneously detect the  $CO_2$  and HDO [21–23], and a coherent DIAL has been demonstrated for measuring  $CO_2$  and wind fields simultaneously [24].

 $CH_4$  has a high global warming potential (GWP) that 27.9 times more potent per kg than  $CO_2$ over 100 years [25]. Monitoring of  $CH_4$  may more urgent due to the dangerous and explosive characteristics, while CH<sub>4</sub> leaks can result in serious and hazardous incidents in some extreme cases that severely jeopardize people's safety [13]. The cost of  $CH_4$  detection equipment is one of the major influences on the ability to fully cover these leak points. Developing innovative technologies to accurately locate and measure CH<sub>4</sub> emissions in an effective and portable manner is extremely important [26,27]. DIAL and IPDA lidar have the advantage of accurately detecting gas concentration with high distance resolution on a large scale of hundreds km<sup>2</sup> [21–24]. However, continuous monitoring of a specific area or a few fixed points using DIAL or IPDA lidar will lead to a higher cost. Tunable diode laser absorption spectroscopy (TDLAS) with simplified structure enables accurate measurement of target gas concentration by rapidly modulating the optical frequency of the laser to cover the specific absorption lines of target gas [28–31]. However, the signal intensity attenuates dramatically with distance is the main reason for limiting long-range detection. Enhancing the response capability of the detector can effectively improve the performance system for path-integrated concentration detection in open path.

Single-photon detectors (SPDs) have the ability to recognize the signal with single-photon intensity, which can overcome the problem that conventional PIN photodetectors cannot recognize weak signals [32]. Superconducting nanowire single-photon detectors (SNSPDs) provides excellent performance of high detection efficiency and broadband response, which have been used for remote sensing [33,34]. However, the practical applications of SNSPDs are limited due to the strict working conditions and the high-cost. Upconversion SPDs exhibit moderate performance and size, but several complex spatial structures are still required [35,36]. Compared with SNSPDs and Upconversion SPDs, InGaAs/InP SPD are more suitable for practical applications with the advantages of small size and low-cost [37,38]. QLM Technology Ltd. has demonstrated a lidar for continuous monitoring of CH<sub>4</sub> based on InGaAs SPD and a technique called TDLidar [39]. Zhu et al. investigated the influences of wedge prism rotation ratios and different environmental conditions on CH<sub>4</sub> leak rate results based on a similar lidar structure [40].

In this work, an all-fiber IPDA lidar with the simplified optical layout is developed for  $CH_4$  leakage monitoring. A homemade InGaAs/InP SPD with multi-channel and multi-mode coupling is used to measure the weak backscattered signal in open path. The intensity and non-linear frequency are corrected for the accurate concentration estimation. A demonstration experiment over ten days is carried out to analyze the accuracy and stability of the system. The preliminary experimental results indicate that the proposed system is reliable for  $CH_4$  continuous monitoring.

# 2. System design and principle

Schematic diagram of the system for CH<sub>4</sub> detection is shown in Fig. 1. Main parameter values of the experimental system are listed in Table 1. The detection light source adopts a continuous wave (CW) distributed feedback (DFB) laser. An arbitrary waveform generator (AWG) provides the modulated signal for driving the laser working in CW scanning mode. The output power of the laser is weakened by a tunable attenuator, avoiding saturation or even damage of the SPD by the return signal with high intensity. The transmitter is a collimator with a diameter of 25 mm. A dynamic gas calibrator is used to mix the gas with a CH<sub>4</sub>/Air ratio of 10<sup>5</sup> ppm. Then, the mixture is flushed into a 40 cm gas cell sealed with optical windows at both ends. The pressure and temperature of the gas cell are 0.16 atm and 298.15 K, respectively. The diffusion medium is a white cement wall at a distance of 12 m from the transmitter. The backscattered signal is

coupled into a multi-mode fiber (MMF,  $50 \,\mu\text{m}$ ) by using another collimator with a diameter of 50 mm, which avoids the interference caused by the strongly mirror reflected signal from the transmitter. A in-line optical filter with a bandwidth of 0.3 nm is used to filter out the background light before the SPD.



**Fig. 1.** Schematic diagram (a) and photo (b) of the experimental system for  $CH_4$  detection. AWG, arbitrary waveform generator; SPD, single photon detector; RM, removable mirror; PD, PIN photodetector; OSC, Oscilloscope; SMF, single-mode fiber; MMF, multi-mode fiber.

Sub-system		Parameter value
	Optical frequency span	181.283 THz ±5 GHz
CW laser	Maximum power	40 mW
	Linewidth	200 kHz
InGaAs/InP SPD	Detection efficiency	5% @ 1650 nm
	DT	200 ns
	AP	~12%
	DC	~500 cps
PIN Photodetector	Detection efficiency	75%
	Dark current	0.1 nA
	Noise Equivalent power	$10^{-15}$ W/Hz <sup>1/2</sup>
Transmitter	Diameter	25 mm
	Focal length	100 mm
Receiver	Diameter	50 mm
	Focal length	200 mm

Table 1. Key parameters of the syste	ameters of the system
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The detection efficiency of the SPD is spectrum-dependent, about 15% at 1550 nm but only 5% at 1650 nm under the same conditions [41]. The dead time (DT), afterpulsing probability (AP) and dark counts (DC) of the SPD are 200 ns,  $\sim$ 12% and  $\sim$ 500 cps, respectively. Multichannel technology is used to increase the maximum count rate of the SPD, which avoiding the

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single-channel SPD tends to operate in the non-linear response region under strong near-field signals and improving the dynamic detection range of the SPD.

A collimator couples the light reflected by a removable mirror into a PIN photodetector and recorded by an Oscilloscope. Figure 2 shows the signals detected by a conventional PIN photodetector including the modulated signal for CW scanning, baseline and the transmission after CH<sub>4</sub> absorption. A square wave signal with a frequency of 100 kHz and a duty cycle of 50% is used as the modulating signal to scan the optical frequency of CW laser up and down across the absorbance spectrum of CH<sub>4</sub> around 181.283 THz [42]. The entire CH<sub>4</sub> absorption cross-section is obtained in both up and down scanning processes.



**Fig. 2.** Signals detected by a PIN photodetector, including the signal for optical frequency tuning, baseline of laser intensity without CH<sub>4</sub> absorption and transmission after CH<sub>4</sub> absorption.

The principle of absorbance spectroscopy is based on the Beer-Lambert law [43,44], which can be expressed as

$$\alpha(v) = -\ln\left(\frac{I_t(v)}{I_0(v)}\right) = S(T)XPL\phi(v),\tag{1}$$

where  $\alpha(v)$  is the absorbance at optical frequency v,  $I_t(v)$  is the intensity of transmission through the gas cell,  $I_0(v)$  is the laser intensity, S(T) is the spectral line intensity related to the temperature T, X is the gas concentration, P is the gas pressure and L is the optical path length of gas absorption.  $\phi(v)$  represents the normalized line profile function at a specific temperature and pressure, which meet the relationship of  $\int_{-\infty}^{\infty} \phi(v) dv \equiv 1$ . According to the temperature and pressure of CH<sub>4</sub> cell, triple-peak Voigt line profile function is used to fit the total absorbance spectrum of the CH<sub>4</sub> at 181.283 THz consists of three absorption lines [21,45], which can be expressed as

$$\phi(v) = \sum_{i=1}^{3} \phi_i(v) = \sum_{i=1}^{3} A_i \frac{2y_i^2}{\omega_{Li} \pi^{3/2}} \int_{-\infty}^{\infty} \frac{e^{-t^2}}{y_i^2 + (x_i - t)^2} dt,$$
(2)

where  $x_i = (4 \ln 2)^{1/2} (v - v_{ci})/\omega_{Gi}$  and  $y_i = (\ln 2)^{1/2} \omega_{Li}/\omega_{Gi}$  are given for simplicity.  $A_i$  is the Voigt area,  $\omega_{Li}$  and  $\omega_{Gi}$  are the Lorentzian full-width at half-maximum (FWHM) and Gaussian FWHM, respectively,  $v_{ci}$  is the center frequency of the absorption line. Mixed absorbance area A can be obtained by integrating the fitted absorbance spectrum, which can be expressed as

$$A = \int_{-\infty}^{\infty} \alpha(v) dv = S(T) X P L.$$
(3)

As a result, path-integrated gas concentration C can be obtained as

$$C = XL = \frac{A}{S(T)P}.$$
(4)

## 3. Experiment

Periodic intensity fluctuation during the scanning is shown in Fig. 2, which can be compensated for by recording the baseline. In addition, the intensity errors caused by the SPD and the non-linear frequency of the laser should be corrected before the concentration estimation. A flowchart of data processing and  $CH_4$  concentration estimation is shown in Fig. 3.



**Fig. 3.** Flowchart of CH<sub>4</sub> concentration detection. *I*, Intensity;  $v_{NL}$ , non-linear frequency; *P*, pressure; *T*, temperature.

Step 1: After obtaining the raw data of the baseline and transmission by SPD. The SPDcorrection is used to sequentially correct the intensity errors caused by DT, AP and DC.

Step 2: The fluctuation of the laser power is compensated by using Eq. (1). Then, the non-linear frequency during CW scanning is measured by homodyne detection and analyzed in joint time-frequency domain.

Step 3: Triple-peak Voigt model is performed to fit the CH<sub>4</sub> absorbance spectra consisting of three absorption lines. According to the temperature and pressure of the CH<sub>4</sub> cell, several HITRAN databased initial conditions are used in the fitting process, including the strength of gas absorption lines, relative frequency positions and FWHM.

Step 4: The path-integrated concentration of  $CH_4$  can be estimated from the aera of the fitted absorbance spectra.

# 3.1. SPD correction

DT, AP and DC are the key parameters of SPDs, and these parameters will be optimized according to the application requirements before actual application. However, due to the interaction between these parameters, photon counts errors still exist under the compromise optimization approach. Details about the SPD correction algorithm can be found in the previous work [46].

Figure 4 shows the baseline of laser intensity and the transmission after  $CH_4$  absorption detected by the SPD before and after correction with time resolution of 1 s. The output power of laser is significantly attenuated to ensure the return signal intensity of 5.4 Mcps during the calibration and demonstration experiments, which allows the SPD operating in the linear response region and guarantees the accuracy of the SPD-correction method. It should be noted that all of the experiments are conducted in an indoor laboratory that the ambient CH<sub>4</sub> concentration is negligible for experimental results. The baseline is measured by directly connecting the fiber into the SPD without the absorption of CH<sub>4</sub>, and the raw data are sequentially corrected for DT, AP and DC, as shown in Fig. 4(a). Figure 4(b) shows the raw data and the SPD-corrected results of the transmission through the  $CH_4$  cell, while the detection distance is 12 m and the output power of the laser is 2 mW. Comparing the raw data and the results corrected by SPD-correction in Figs. 4(a) and 4(b), DT and AP cause a large error on the photon counts, while the influence caused by DC can be neglected. The CH<sub>4</sub> absorbance spectrum detected by PIN photodetector is used as a reference for comparison, as shown in Fig. 4(c). The SPD-corrected absorbance curve resulted in significantly higher absorbance compared to the raw data, specifically the top point of CH<sub>4</sub> absorbance is 0.77 and 0.92 before and after correction. The absorbance after SPD-correction is in high agreement with the results detected by PIN photodetector, which indicates that the SPD-correction method effectively corrects the intensity errors caused by DT, AP and DC on photons counts. Figure 4(d) shows the differences of absorbance measured

by SPD before and after correction in contrast to the curve measured by PIN photodetector. In the larger portion of the absorbance as indicated by yellow shading areas in Figs. 4(c) and 4(d), the maximum difference between SPD raw data and PIN photodetector-measured one is larger than 16.4% while the mean difference is 4.95%. After SPD-correction, the maximum difference and mean difference can be decreased to 2.6% and 0.84%. Note that, the residual relative differences after SPD-correction show symmetric distribution about the centers. As we pointed out earlier, this phenomenon is due to the different sampling methods between the digital and analog detection [47], where the SPD averages the photon counts over a period of time (50 ns), while the PIN photodetector samples the signal at 1 GSa/s.



**Fig. 4.** (a) Laser baseline and (b) transmission signal detected by SPD and sequentially corrected for DT, AP, and DC. (c) Comparison of  $CH_4$  absorbance detected by PIN photodetector and SPD. (d) Given the results measured by PIN PD as a reference, the differences of absorbance using SPD before and after SPD-correction. Diff.: difference.

## 3.2. Non-linear frequency correction

The modulating signal drives the current fed into the laser, affecting the temperature and the carrier concentration in the gain medium, both of which will change the optical frequency of laser [48]. The carrier concentration in the active layer of the laser increases rapidly with a step increase of the modulated current, resulting in an accelerated change in optical frequency and gradually slows down to a steady-state value due to thermal effects [49]. As a result, the gas absorption line profile is distorted, since the optical frequency of laser varies nonlinearly versus time during scanning.

An unbalanced Mach-Zender interferometer (MZI) is served as a frequency discriminator to measure the non-linear frequency scanning [50], as shown in Fig. 5. A CH<sub>4</sub> cell with a fiber pigtail is connected in front of the MZI, and the position of the CH<sub>4</sub> absorption peak can be found. This position will be used as a reference for absolute optical frequency for correcting non-linear frequency. The unbalanced MZI consists of two 50:50 couplers (C1, C2) and a length of delay line (the length difference is 1 m between the two arms). The beat frequency is measured

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by a PIN photodetector with a bandwidth of 1 GHz and recorded by an OSC at sampling rate of 1 GSa/s.



Fig. 5. Schematic of the unbalanced MZI for homodyne detection. C, Coupler.

The time delay introduced in two arms of MZI is ~5 ns that is much smaller than the scanning period of 5 µs, thus the beat frequency f(t) can be expressed as the time derivative of optical frequency

$$f(t) = \frac{dv(t)}{dt}\tau,$$
(5)

where v(t) is the instantaneous optical frequency of laser during CW scanning,  $\tau$  is the time delay difference introduced in MZI. Joint time-frequency analysis (JTFA) maps a two-dimensional time function into a three-dimensional time-frequency distribution to characterize the intensity of signals at different times and frequencies, which has been widely applied in the fields of machinery inspection [51], seismic exploration [52] and lidar [53]. Based on advantage of the refined processing for signal in time and frequency, JTFA is used to process the beat frequency to obtain the instantaneous optical frequency variation. According to the optical frequency of the gas absorption peak and corresponding time position, the exact instantaneous optical frequency can be estimated by integrating the beat frequency, which is given as

$$v(t) = v_0 \pm R \int_{t_0}^t f(t) dt,$$
(6)

where  $v_0$  is the optical frequency of the gas absorption peak. In this work R = 0.2 is the ratio coefficient of sampling interval  $t_{sa}$  and delay time  $\tau$ . Different from the conventional interferometric approaches only the frequency information at the interferometric peak or valley positions can be obtained, the instantaneous frequency change at each sampling point can be estimated by analyzing the beat frequency in joint time-frequency domain. As a result, more details of the frequency variations can be estimated by using JTFA thus enabling more accurate non-linear frequency corrections.

Figure 6 illustrates the beat frequency signals and the results processed by JTFA. Figure 6(a) is the time-domain interferogram of two same CW scanning signal obtained by homodyne detection with different delay time, and the absorption information of  $CH_4$  is contained within the intensity of beat frequency signal. The effects of high-frequency and low-frequency noises are eliminated by a band-pass filter. The filtered beat frequency is processed by the JTFA with Choi-Williams distribution (CWD), then the beat frequency f(t) i.e. instantaneous optical frequency change can be obtained, as shown in Fig. 6(b). The black solid line represents the maximum intensity of beat frequency signal at the corresponding time sampling point, the corresponding vertical coordinate is the instantaneous optical frequency change. It can be clearly seen that the optical frequency of the laser varies nonlinearly with time, accelerating at the instant when the high and low level of modulating signal changes and slowing down at the point where closes to the CH<sub>4</sub> absorption peak. As a result, the absorbance spectrum in the time domain exhibits a distinctly asymmetric structure, as shown in Figs. 7(a) and 7(b). The total optical frequency range of CW scanning and the real optical frequency can be obtained by integrating the beat frequency at every time sampling points according to Eq. (6). Figure 6(c) shows the real optical frequency v(t) during CW scanning. The optical frequency span in the scanning is about 10 GHz.



**Fig. 6.** (a) Temporal interferogram. (b) Beat frequency f(t) processed by the JTFA with CWD. (c) Optical frequency v(t) during CW scanning.



**Fig. 7.** The absorbance spectra before and after non-linear frequency correction in (a) down and (b) up scanning. (c) Comparison of absorbance spectra in up and down scanning. The subfigure below (c) is the difference between the both absorbance spectra.

Figures 7(a) and 7(b) show the absorbance spectra before and after non-linear frequency correction in down and up scanning, respectively. A symmetry-calibration process that comparing the absorbance spectra of up and down scanning is used to verify the accuracy the correction at this step, as shown in Fig. 7(c). The maximum difference is less than 1% with mean difference of 0.33% is achieved within a span of 4 GHz around the center of absorbance spectrum, which indicating that the non-linear frequencies during scanning are well corrected.

# 3.3. Accuracy and stability

After correcting for intensity errors due to photon counts and non-linear frequencies during scanning, triple-peak Voigt model is used in the fitting process of  $CH_4$  absorbance spectra nearby 181.283 THz consists of three absorption lines. The final absorbance spectra used for fitting are the average of both spectra in up and down scanning. The Levenberg-Marquardt method is used to achieve the numerical least-squares optimization of the Voigt fitting.

Figure 8(a) shows the ten typical triple-peak Voigt fitting results of  $CH_4$  absorbance spectrum at a detection distance of 12 m, while the signal intensity is 5.4 Mcps and the time resolution is 1 s. The absorbance spectra corresponding to the three absorption lines and the mixed one show a high degree of agreement at strong signal intensity. The standard deviation of residual

between the corrected results and fitting results is 0.4%. The backscattered signal received in the frequency of *v* and at a range of *R* can be expressed by [54]

$$P(v,R) = \frac{K\beta(v,R)T_{r}^{2}(v,R)}{R^{2}},$$
(7)

where *K* is the constant of lidar system,  $\beta(v, R)$  is the volume backscatter coefficient,  $T_r(v, R)$  is the transmittance of atmosphere. According to the Eq. (7), the backscattered signal through the atmosphere decays proportionally to the square of the distance. At the real detection distance of 12 m, the laser output power is attenuated by 20 dB from 2 mW to 0.02 mW, which leading the signal intensity decreasing to 54 kcps. This signal intensity corresponds to the detection distance of 120 m. Figure 8(b) shows the triple-peak Voigt fitting results of the absorbance spectra, in which the weak signals lead to large fluctuation in the residual curves with a maximum of 8.51%.



**Fig. 8.** Results of ten typical tests after triple-peak Voigt fitting at two different signal intensities with a time resolution of 1 s, (a) signal intensity of 5.4 Mcps with the detection distance of 12 m, (b) signal intensity of 54 kcps. The subfigures below (a) and (b) are residuals between the results of experiment and fitting. t, test.

To further validate the accuracy and stability of the system, continuous observations over 10 days are carried out at two different signal intensities. Figure 9 shows the 40 groups of test results both in the intensity of 5.4 Mcps and 54 kcps, while the test is carried out every 3 hours and each group of tests contain 20 results. The concentration of the  $CH_4$  cell is  $10^5$  ppm with the optical path of 40 cm, which corresponding path-integrated concentration is  $4 \times 10^4$  ppm·m. Figure 9(a) shows the path-integrated concentration measured at a signal intensity of 5.4 Mcps with a time resolution of 1 s. The measurements results show good consistency, where mean deviation of 10.47 ppm·m (0.03%) with standard deviation of 182.22 ppm·m (0.46%) is achieved. Figure 9(b) shows the results measured at the signal intensity about 54 kcps at a time resolution of 1 s. The mean deviation and the standard deviation are degraded to  $529.04 \text{ ppm} \cdot \text{m} (1.32\%)$ and 1732.27 ppm·m (4.33%), respectively. The larger concentration deviation is due to the lower signal-to-noise ration of the weak signal intensity, and the results in weak signal intensity can be used to simulated the return signal from the detection distance of 120 m or the objects with weak reflection coefficient. It should be noticed that the relative output power is only the 1/20 of the maximum power, which indicates that the accuracy can be improved by increasing the laser power in field test.

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**Fig. 9.** Continuous observation over 10 days of  $CH_4$  path-integrated concentration detection. (a) Detection distance of 12 m, with the signal intensity of 5.4 Mcps and a time resolution of 1 s. (b) Simulated detection distance of 120 m by attenuating the signal intensity to 54 kcps with a time resolution of 1 s. The time interval between adjacent tests is 3 hours and each test contained 20 results.

# 4. Conclusion

An IPDA lidar with simplified all-fiber optical layout are demonstrated for  $CH_4$  leakage monitoring. For the accurate concentration estimated, the intensity is corrected by SPD-correction and the non-linear frequency is corrected by homodyne detection and JTFA. The accuracy and stability are verified by a continuous observation over ten days. A mean deviation of 0.03% with standard deviation of 0.46% is achieved at a distance of 12 m and a time resolution of 1 s. The detection performance with weak signal intensity that attenuated by 20 dB is verified with a mean deviation of 1.32% and a standard deviation of 4.33%.

The preliminary experiment results demonstrated that the proposed all-fiber IPDA lidar system for  $CH_4$  detection is reliable. In the near future, several measures will be applied to further enhance the performance of system. The linear frequency variation during CW scanning can be realized by using pre-distortion for modulated current [55,56], which will reduce the errors caused by non-linear frequency variations. The distance to the target can be realized by integrating a small-sized and low-cost ranging lidar, and more optical power can be used for improve the detection distance. The laser, SPD, transmitter and receiver framed by the white dashed line in Fig. 1(b) will be integrated, thus the system can be used as a portable lidar. In addition, benefiting from the wide response of InGaAs/InP SPD in the near-infrared band, the improved system can be adapted to the continuous monitoring of other gas leaks, such as H<sub>2</sub>S, CO, NH<sub>3</sub>, by integrating more laser sources using optical switches.

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