Recent Advances in Light-Induced Thermoelastic Spectroscopy for Gas Sensing: A Review

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Abstract: Light-induced thermoelastic spectroscopy (LITES) is a promising optical approach for gas sensing, which uses a quartz tuning fork (QTF) as a photothermal detector, instead of a commercial photodetector. Since the QTF has the advantages of low cost, small size, high resonance frequency, high-quality factor (Q-factor), and a wide spectral response range, and the LITES sensor has received extensive attention and obtained great development. This review paper summarizes and discusses the advances of the QTF-based, state-of-the-art LITES gas sensing technique in recent years and presents the development prospects of LITES sensor in the future.

Keywords: light-induced thermoelastic spectroscopy; gas detection; optical sensor; quartz tuning fork

1. Introduction

Trace gas detection plays an important role in many fields, such as environmental monitoring, power industry, industrial process control, noninvasive medical diagnosis, life science, and so on. Optical gas sensors provide an effective solution for trace gas detection, due to their merits of high selectivity and sensitivity, fast response, cross-interference free, and in situ detection ability [1–29].

Quartz tuning fork (QTF), a device with piezoelectric effect, has been applied in quartz-enhanced photoacoustic spectroscopy (QEPAS) gas sensing technique since 2002 [30–44]. The QTF has the advantages of a low cost (<$1), a small size (<0.3 mm$^3$), a high resonance frequency (~32.768 kHz), and a high Q-factor (~100,000 in vacuum, ~10,000 at atmospheric pressure). In QEPAS, the QTF was used as a resonant acoustic-electric transducer to detect the photoacoustic signal, instead of a photoacoustic cell and a microphone in conventional photoacoustic spectroscopy (PAS) [45–65]. The QEPAS-based gas sensor has an immunity to ambient acoustic noise and the capability to analyze ultra-small volume gas samples [54–65]. However, the QTF must be in contact with the target gas; thus, the performance of the QEPAS sensor can be deteriorated when it is applied in some specific conditions, such as combustion diagnostics and corrosive gas detection.

Light-induced thermoelastic spectroscopy (LITES) is a novel QTF-based gas sensing technique, which was proposed by Ma et al. in 2018 [66]. A schematic diagram of light-induced thermoelastic spectroscopy is shown in Figure 1. In LITES, the QTF was employed to measure the variation of the light intensity absorbed by the target gas. The modulated light beam, after being absorbed by the target gas, irradiates the surface of the QTF. The QTF’s thermal energy generated by light absorption is transformed into the mechanical vibration of the QTF via the light-thermo-elastic conversion. The mechanical vibration can be enhanced when the QTF operates in the resonance mode. Based on its piezoelectric effect, the QTF converts the mechanical vibration into an electrical signal that contains the information of target gas concentration [39,66–70]. LITES is also called...
quartz-enhanced photothermal spectroscopy (QEPTS). Compared with the traditional photodetector-based direct absorption spectroscopy (DAS) sensor, the QTF-based LITES sensor has a low cost and a small size and can operate in a wide spectral range without any cooling systems [39,66–84]. In a LITES sensor, the QTF does not require contact with the target gas. Therefore, the LITES technique can achieve the non-contact remote and standoff gas detection, which is different from the QEPAS technique.

![Figure 1. The schematic diagram of light-induced thermoelastic spectroscopy.](image)

In this review, we summarized the research advances in the LITES sensing technique in recent years and gave the prospects for the future. We divided the LITES sensors reported in the literature into six types, based on the sensor system structure and the QTF type. Conventional LITES sensor, combination of QEPAS and LITES, and multi-QTF-based LITES sensor have three sensor structures with different QTF configurations, respectively. Material-coated QTF and custom QTF are two different types of QTF, which were exploited to improve the light response sensitivity, compared with the commercial standard QTF. Optical fiber-based LITES sensors used optical fiber as a gas cell, used optical fiber to couple a free-space gas cell, or used optical fiber to measure the vibration of QTF, which can make full use of the advantages of optical fiber.

2. Conventional LITES Sensor

The conventional LITES sensor uses a commercially available QTF, without any modifications, to detect the absorption spectrum signal of the target gas [66,67,85–98]. The resonance frequency and Q-factor of commercial standard QTFs are typically ~32.768 kHz and ~10,000 at atmospheric pressure. Due to slight structural differences, the resonance frequency and Q-factor of different commercial QTFs are also different. The first LITES sensor was reported by Ma et al. in 2018 [66], which used a 20 cm gas absorption cell and a commercial QTF with a resonance frequency of 30.72 kHz to detect C2H2. A 1.53 μm distributed feedback (DFB) diode laser was selected as the excitation source. The experimental setup is shown in Figure 2. After passing through the gas absorption cell, the laser beam was focused onto the surface of the used QTF. By using the finite element method (FEM) and simulation tool COMSOL Multiphysics software, it can be seen that the incident position of the laser beam on the QTF can affect the signal level, which can support the experiment results. Moreover, a signal enhancement by increasing the laser excitation power was observed in the experiment. A 1σ minimum detection limit (MDL) of 718 ppb was obtained, with an integration time of 1 s, corresponding to the normalized noise equivalent absorption (NNEA) coefficient of 7.63 × 10⁻⁹ cm⁻¹ W/√Hz. The MDL is better than the reported result of 200 ppm, based on tunable diode laser absorption spectroscopy (TDLAS) in Ref. [99], and the NNEA is better than the reported result of 3.54 × 10⁻⁸ cm⁻¹ W/√Hz, based on QEPAS in Ref. [100].

In 2019, He et al. reported an ultra-highly sensitive LITES sensor for CO detection at 2.23 μm [67], which combined a high Q-factor QTF and a multipass cell with an optical pathlength of 10.1 m to improve the performance of the LITES sensor. The experimental setup of such a LITES sensor system is shown in Figure 3. The optimization for the position of the laser focal point on the QTF’s surface was simulated by using COMSOL Multiphysics software, and the optimum laser focal point was determined at the bottom of the QTF’s prongs to obtain the strongest LITES signal, as shown in Figure 3d. In Ref. [66], due to the limited investigation of the QTF, the applied position was located on the edge of the prong. The resonant frequency of the used QTF slightly increased from 30,711.4 to 30,717.2 Hz.
when the pressure was from 760 to 20 Torr. However, the Q-factor increased significantly with decreasing pressure, and a Q-factor of 50,177 was obtained at 20 Torr, which showed a 3.4 times improvement, compared with an atmospheric pressure. A 1σ MDL of 470 ppb was achieved for a 60 ms integration time at 20 Torr. The corresponding noise equivalent absorption (NEA) coefficient and NNEA coefficient were $2.0 \times 10^{-7}$ cm$^{-1}$/√Hz and $7.4 \times 10^{-10}$ cm$^{-1}$/W/√Hz, respectively. They confirmed the better performance of LITES, with respect to TDLAS, by a comparative experiment under the same conditions.

![Diagram of QTF setup](image)

**Figure 2.** Experimental setup of the LITES sensor system. CW-DFB diode laser: continuous wave, distributed feedback diode laser; FC: fiber collimator; W: CaF$_2$ window; L: CaF$_2$ plano-convex lens; TA: transimpedance amplifier; ADC: analog-to-digital converter. Reproduced with permission from Ref. [66]. Copyright 2018, copyright The Optical Society.

![Diagram of gas detection](image)

**Figure 3.** (a) Experimental setup of the LITES sensor system. (b) The specific optical path structure. (c) The image of a QTF. (d) Stimulated normalized integration for thermoelastic induced mechanical displacement of the QTF’s surface. L1: lens 1; FC: fiber collimator; QTF: quartz tuning fork; PD: photoelectric detector; CW-DFB Laser: continuous wave distributed feedback laser; PC: personal computer; MFLI: the model of the used lock-in amplifier. Reproduced with permission from Ref. [67]. Copyright 2019, copyright The Optical Society.

To improve the practical application ability of the LITES sensor, it is necessary to make the sensor highly sensitive and compact. The miniaturized multipass cell can provide a way for an effective solution to realize the compact LITES sensor. In 2021, Wei et al. developed a palm-sized LITES sensor to detect CH$_4$ at 1.65 μm [85]. A fiber-coupled mini-multipass cell with an optical pathlength of 4.2 m and a commercial standard QTF composed a gas detection module with a compact dimension of 78 mm × 40 mm × 40 mm by using 3D...
printing technology. The integrated design made the sensor have excellent robustness and stability. The resonance frequency and $Q$-factor of the used standard QTF at 8 Torr were 32,780.9 Hz and 27,205, respectively. An MDL of 52 ppb was achieved for a 300 ms integration time when the QTF operated at 8 Torr, corresponding to the NNEA coefficient of $2.1 \times 10^{-8}$ cm$^{-1}$ W/$\sqrt{\text{Hz}}$. The experimental result is better than the CH$_4$ TDLAS sensor, with an MDL of 117 ppb reported in Ref. [101]. Such a CH$_4$ LITES sensor was applied for a seven-day continuous monitoring of atmospheric CH$_4$ concentration, verifying the long-term stability of the sensor.

The application of LITES sensor in multi-gas detection was also greatly developed. In 2020, Xu et al. reported a multi-gas LITES sensor based on frequency-division multiplexing [86], which employed a single commercial QTF with a resonance frequency of 32,781.73 Hz and three near-infrared DFB diode lasers to simultaneously detect H$_2$O, CO$_2$, and CH$_4$. The three-laser beam was coupled by a 3 $\times$ 1 fiber coupler and collimated to pass through the gas absorption cell by a collimator. Finally, the coupled laser beams were focused onto the surface of the QTF to excite the LITES signal. Since the 2$^f$ detection technique in the experiment, the laser for H$_2$O detection was modulated at 16,390.5 Hz, which was the half of the QTF’s resonance frequency. The other two lasers were modulated at 16,391.0 Hz and 16,391.5 Hz for CO$_2$ detection and CH$_4$ detection, respectively. There was a frequency interval among the modulation frequencies of three lasers, which were in the resonance bandwidth of the QTF. This method cannot make the QTF obtain the strongest resonance response when measuring each gas, but it can achieve multi-gas simultaneous detection by using a single QTF in the LITES sensor.

3. Combination of QEPAS and LITES

Both QEPAS and LITES are QTF-based laser absorption spectroscopy. If the QEPAS and LITES techniques are combined, the sensor performance can be further improved by adding the QEPAS and LITES signals [102–107]. In 2021, Hu et al. demonstrated a two-QTF-based QEPAS-LITES sensor system for H$_2$O detection [102], as shown in Figure 4. The two QTFs, with a resonance frequency difference of ~0.4 Hz, were used to detect QEPAS and LITES signal, respectively. The laser beam passed through the prongs gap of the QTF1 to excite QEPAS signal, and then was focused on the bottom of the QTF2’s prongs to excite LITES signal. The measured 1.01% H$_2$O signals of QTF1, QTF2, and QTF1 + QTF2 for QEPAS, LITES, and QEPAS-LITES were 9.34 $\mu$V, 88.27 $\mu$V, and 95.25 $\mu$V, respectively. So, the QEPAS-LITES signal was 10.2 times higher than the QEPAS signal and 1.1 times higher than LITES signal. Due to the difference of resonance frequency between the two QTFs, the response for QTF1 and QTF2 cannot be the strongest simultaneously under the same modulation frequency. So, the signal of QTF1 + QTF2 was not equal to the sum of the signals of QTF1 and QTF2. The measured noise levels of QTF1, QTF2, and QTF1 + QTF2 were 31.81 nV, 66.46 nV, and 70.24 nV, respectively. The noise levels of LITES and QEPAS-LITES sensor system were higher than that of QEPAS, which was mainly attributed to the thermal noise of QTF. The suppression methods of system noise in LITES sensor were studied in Refs. [108,109]. To avoid resonance frequency mismatching of the QTFs reducing the sensor performance, Ma et al. proposed two types of resonance frequency matching methods, based on temperature regulation and pressure regulation, in 2022 [110].

The simultaneous measurement of the QEPAS signal and LITES signal based on single QTF has also been also studied [104–107], which does not have the problem of the resonant frequency mismatching of QTFs in the two-QTF-based QEPAS-LITES sensor. In 2019, Zheng et al. used a QTF and a 1.368 $\mu$m DFB laser, with a power of ~10 mW, to detect H$_2$O in air [104]. The laser beam illuminated the QTF prong spacing and the prong surface; thus, the QTF generated the QEPAS signal and LITES signal simultaneously. They called this method quartz-enhanced photothermal-acoustic spectroscopy (QEPTAS), as shown in Figure 5. When the QTF was located at distances of 1 m and 2 m from the laser head, the MDLs of ~12 ppm and ~55 ppm were obtained, corresponding to the NNEA coefficients of $8.4 \times 10^{-7}$ cm$^{-1}$ W/$\sqrt{\text{Hz}}$ and $3.7 \times 10^{-6}$ cm$^{-1}$ W/$\sqrt{\text{Hz}}$, respectively. Xu et al. also
reported a similar technique by using an external cavity quantum cascade laser (ECQCL) in 2020. In 2021, Qiao et al. realized the simultaneous measurement of the QEPAS signal and LITES signal by using a QTF and a right-angle prism \[^{[105]}\], as shown in Figure 6. They called this method single-quartz-enhanced dual spectroscopy (S-QEDS). The laser beam was firstly collimated to pass through the QTF prongs’ spacing and then was reflected by the right-angle prism to incident on the bottom of the QTF’s prongs. For a 0.58% H\(_2\)O, the QEPAS signal and LITES signal were 5.49 \(\mu\)V and 16.12 \(\mu\)V, respectively. The S-QEDS signal was 21.53 \(\mu\)V, which almost equal to the sum of QEPAS signal and LITES signal.

Figure 4. Experimental setup of the QEPAS-LITES sensor system. \(\Sigma\): adder. Reprinted/adapted with permission from Ref. \[^{[102]}\]. Copyright 2021, copyright The Optical Society.

Figure 5. Experimental setup of the QEPTAS sensor system. Reprinted/adapted with permission from Ref. \[^{[104]}\]. Copyright 2019, copyright MDPI.
Figure 5. Experimental setup of the QEPTAS sensor system. Reprinted/adapted with permission from Ref. [104]. Copyright 2019, copyright MDPI.

Figure 6. (a) S-QEDS sensor system configuration. (b) QEPAS sensor system configuration. (c) LITES sensor system configuration. QTF: quartz tuning fork; FC: fiber collimator; PC: personal computer. Reprinted/adapted with permission from Ref. [105]. Copyright 2021, copyright The Optical Society.

4. Multi-QTF-Based LITES Sensor

In 2015, Ma et al. reported multi-quartz-enhanced photoacoustic spectroscopy (M-QEPAS) [111], which used two QTFs to achieve a 1.7 times signal enhancement with respect to the single QTF-based QEPAS method under the same operating conditions. Similarly, they extended this idea to LITES. A multi-QTF-based LITES sensor system was demonstrated by them in 2020 [112]. The experimental setup was shown in Figure 7. The metal film with high reflectivity at the optimal laser focusing position of the QTFs was removed, in order to improve the laser absorption and transmission. The detailed etching method can be obtained from Ref. [112]. A DFB diode laser with output power of 10.62 mW was used to detect C\textsubscript{2}H\textsubscript{2} at 1530.37 nm. The collimated laser beam passed through the gas absorption cell and was focused on the bottom of the two prongs of QTF1. Then, the laser beam transmitted the QTF1 that had a transmission of ~60% for 1.53 µm laser after etching and was focused the bottom of the two prongs of QTF2. The measured 2% C\textsubscript{2}H\textsubscript{2} signals of QTF1, QTF2, and QTF1 + QTF2 were 650.05 µV, 372.56 µV, and 983.44 µV, respectively. Due to the QTF1 transmission of ~60% for 1.53 µm laser, the signal of QTF2 was 57% of QTF1. The difference of resonant frequency between the two QTFs resulted that the signal of QTF1 + QTF2 was not equal to the sum of the signals of QTF1 and QTF2, which was similar to the two-QTF-based QEPAS-LITES sensor [102] mentioned in Section 3 and the two resonance frequency matching methods proposed in Ref. [110] can also be applied to the two-QTF-based LITES sensor. So the two-QTF-based LITES sensor achieved a 1.51 times signal enhancement compared with the single etched QTF1 based LITES sensor. In addition, the experimental results showed that the etched QTF1 achieved a ~1.30 times signal enhancement compared with the standard QTF1 without etching. The noise level was measured when the absorption cell was filled with pure N\textsubscript{2}. The measured 1σ noise levels of QTF1, QTF2 and QTF1 + QTF2 were 47.79 nV, 47.57 nV, and 47.80 nV, respectively, which had no obvious difference. Based on the measured results, the two-QTF-based LITES sensor obtained an MDL of 0.97 ppm for C\textsubscript{2}H\textsubscript{2} detection with an integration time of 1 s and the MDL can be further improved to 0.19 ppm, with an optimum integration time of 200 s, by using an Allan variance analysis.
The QTF’s piezoelectric signal is related to the light energy absorbed by the QTF in LITES sensor. However, the silver film with high reflectivity coated on a commercial standard QTF’s surface can minimize the laser absorption of the QTF, thus reducing the performance of LITES sensor. In Ref. [112], mentioned in Section 4, it was demonstrated that the LITES signal generated by the QTF without silver film was ~1.30 times stronger than that with silver film. Therefore, some suitable materials were considered to be coated on the commercial QTF’s surface to increase the absorption of light and the efficiency of photothermo-elastic-electric conversion, which can effectively enhance the LITES signal [113–119]. In 2021, Zhou et al. designed a QTF (also called quartz crystal tuning fork, QCTF) with an ultrathin iron-doped cobaltous oxide (Fe–CoO) coating [113], as shown in Figure 8, which enhanced the detection performance by a factor of 4.5, due to an extremely large specific surface area and a high charge migration rate of the Fe–CoO film. Soon afterwards, a graphene-coated QTF was reported for LITES sensor by Lou et al. [114]. The charge-transfer enhancing effects of graphene made the graphene-coated QTF-based LITES sensor obtain a gain factor of 1.8 and 1.7 for the sensitivity and signal-to-noise ratio (SNR), respectively, compared with the conventional LITES sensor, based on a bare QTF without coating. In addition, the strong UV to THz light absorption of graphene offers a promising strategy for sensitive trace gas detection in an ultra-broadband spectral range. They also reported a polymer-coated QTF for enhancing the sensitivity of LITES, due to the large thermal expansion coefficient of polymer [115]. The detailed theory and material properties can be found in Ref. [115]. The experimental results showed that the polymer-coated QTF-based LITES sensor achieved a maximum gain factor of 3.46 and 3.21 for the signal amplitude and SNR, respectively. In 2022, carbon nanotubes (CNT), graphene oxide (GO), and reduced graphene oxide (rGO), the three nanomaterials with high light absorption coefficient, were coated on QTF’s surface by Wang et al. [118] and obtained the LITES SNR gain factors of 1.54, 1.33, and 2.24, respectively, with respect to the bare QTF. The reported results of these references mentioned above can confirm that modifying the coating on the QTF’s surface by use of suitable materials is an effective method for improving the performance of LITES sensor, but in this case, a complex process is required.
enhanced the detection performance by a factor of 4.5, due to an extremely large specific surface area and a high charge migration rate of the Fe–CoO film. Soon afterwards, a graphene-coated QTF was reported for LITES sensor by Lou et al. [114]. The charge-transfer enhancement effects of graphene made the graphene-coated QTF-based LITES sensor obtain a NNEA coefficient of 9.16 × 10−10 cm−1 W/√Hz, which is ~10 times better than a standard 32.768 kHz QTF-based LITES sensor. Soon afterwards, Russo et al. evaluated the performance of five custom QTFs and a standard QTF for LITES sensor in detail [127]. It is worth mentioning that their research group has designed a lot of custom QTFs with excellent performance for QEPAS sensors [36,120,123,130–132]. The characteristic parameters of the used six QTFs are shown in Table 1. The detailed structure and size of the five custom QTFs can be found in Ref. [127]. They obtained the conclusion that the LITES SNR is proportional to the product of the strain and the QTF accumulation time, and the LITES SNRs of the five custom QTFs are higher than that of the commercial standard QTF, as shown in Figure 10, which was explained in detail in the paper. The custom QTF#2 had the highest SNR. A comparative experiment showed that the SNR of the QTF#2 at 5 Torr was ~6.5 times higher than that of a commercial amplified near-infrared photodetector.

**Table 1.** The characteristic parameters of the used six QTFs. $f$: measured resonance frequency of the fundamental mode; Q-factor: related quality factor; $\tau$: accumulation time; $\epsilon$: maximum strain field. Reproduced with permission from Ref. [127]. Copyright 2020, copyright The Optical Society.

<table>
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In 2021, Wei et al. systematically studied the response characteristics of the custom QTF in a wide spectral range of 1.65–10.34 µm, based on a LITES sensor system [70]. The custom QTF#2 in Ref. [127], mentioned above, was used for the experiment, due to its excellent SNR. They measured the transmittance spectrum of the QTF quartz by using a Fourier transform spectrometer, as shown in Figure 11. A high transmissivity of >90% was obtained in 1–3.5 µm, and then it rapidly decreased in 3.5–4.8 µm. The narrow absorption features around 2.7 and 4.2 µm were due to H₂O and CO₂ absorption, respectively. When the wavelengths were >4.8 µm, the transmissivity reached values close to zero. To achieve a high thermoelastic conversion efficiency in the whole infrared spectral range, a chromium film with the feature of strong absorption in the mid-infrared range and a gold layer with a high reflectivity were coated on the QTF’s back surface, as the front surface was uncoated. The chromium film was coated between quartz crystal layer and gold layer as an excellent adhesion promoter. The laser was back-reflected at the chromium–gold interface to enhance the light absorption. FEM analysis was used to relate the energy release with the induced thermal distribution, as described in detail in Ref. [70]. In the experiment, the measurements were based on a LITES sensor system by using five single-mode DFB laser sources covering the infrared range from 1.65–10.34 µm. An average responsivity of ~2.2 kV/W and noise-equivalent power (NEP) of 1.5 nW/√Hz were achieved for the custom QTF as a photothermal detector, which can be flatly extended across the whole

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infrared spectral range of 1.65–10.34 μm, but without requiring any thermoelectrical cooling system. The responsivity comparison between the used custom QTF and commercial photodetectors is shown in Figure 12. Finally, they demonstrated a heterodyne detection method in the LITES sensor system to measure the resonance properties of the QTF, as well as the target gas concentration in the absorption cell, similar to the approach proposed for QEPAS in Refs. [35,64]. The combination of heterodyne detection method and LITES technique can avoid the frequency calibration of the QTF in practical application. Further, Ma et al. recently reported a high-robustness, self-correlated, heterodyne-based LITES sensor system, which realized the compensation of the measurement errors caused by laser power variation, focus position shift, and QTF resonant frequency shift [96].

![Figure 11](image1.png)

**Figure 11.** Continuous line: transmittance spectrum of the QTF quartz measured using a Fourier transform spectrometer. Dashed lines: the central wavelengths of the used lasers in this work. Reprinted/adapted with permission from Ref. [70]. Copyright 2021, copyright AIP Publishing.

![Figure 12](image2.png)

**Figure 12.** Solid lines: responsivity as a function of the wavelength for three models of photovoltaic Vigo detectors, namely PVI-4 (red line), PVI-6 (blue line), and PVI-10.6 (green line) HgCdTe, and for Thorlabs PDA10CF InGaAs Amplified Photodetector (black line), as provided by the device manufacturers. Black square: responsivity as a function of the wavelength for the used custom QTF. Reprinted/adapted with permission from Ref. [70]. Copyright 2021, copyright AIP Publishing.
7. Optical Fiber-Based LITES Sensor

Optical fiber has the advantages of small volume, light weight, long transmission distance, anti-harsh environment, immunity to electromagnetic interference, easy formation of sensor networks, etc. The application of optical fiber in LITES sensor can greatly improve its performance and practicability [69,133–138]. In 2020, Hu et al. developed an all-fiber LITES sensor system [69], as shown in Figure 13. A hollow-core photonic crystal fiber (HC-PCF), with a length of ~30 cm, was used to confine the gas sample and transmit light modes simultaneously, instead of a multipass cell. Moreover, the light signal with gas absorption was guided on the QTF’s surface by a single-mode optical fiber (SMF) tip, rather than an optical lens. They found that the SMF tip should be as close to the QTF’s surface as possible to improve the detection performance, since a smaller beam waist radius and less laser radiation loss in the air can be achieved. A 1653.72 nm DFB diode laser was used as the excitation source for CH₄ detection. They obtained a NNEA coefficient of 9.66 × 10⁻⁹ cm⁻¹ W/√Hz, which was similar to a free-space LITES sensor. The performance of the all-fiber LITES sensor would be further improved if the mode interference noise was suppressed effectively. In 2022, Ma et al. and Boješ et al. both reported a similar experimental scheme [133,134], which employed a hollow-core anti-resonant fiber as a gas absorption cell.

In 2021, Hu et al. proposed another optical fiber-based LITES sensing method [135], which used a fiber-coupled sensing probe, as shown in Figure 14a. The fiber-coupled structure was realized by two SMFs (input and output) and two FC/APC (Ferrule contactor/angle physical contact) connectors with a 0.25 dB connection loss. The experimental setup of the fiber-coupled sensing probe-based LITES sensor system is shown in Figure 14b. They exploited a near-infrared DFB diode laser to detect CH₄ at 1653.72 nm. The incident laser from the input SMF was collimated by a fiber-coupled collimator and reflected by a reflector to another fiber-coupled collimator. Then, the laser was coupled to the output SMF, and finally, focused on the QTF’s surface by a fiber-coupled focuser. The distance between the collimators and the reflector was ~15 cm, resulting in a total optical pathlength of ~30 cm. The sensor probe was placed outside the laboratory, ~100 m away, by using two ~100 m long SMFs. They achieved the long-distance in situ monitoring of CH₄ leakage in campus by employing such a fiber-based LITES sensor. Subsequently, they used a similar fiber-coupled structure combined with a fiber-coupled off-axis cavity and a 1512.24 nm DFB laser to realize in situ multipoint NH₃ leak monitoring [136]. Fiber-coupled multipass...
cell-based LITES sensor systems with a similar experimental scheme were also reported in recent years [129,137].

In 2022, Pan et al. combined the highly sensitive fiber optic Fabry–Pérot interferometry (FPI) technique with LITES to develop an all-optical LITES sensor system [138], which exploited the FPI method to measure the vibration of the QTF, instead of the conventional piezoelectric detection, as shown in Figure 15. The end surface of the optical fiber and the silver-coated surface of the QTF’s prong formed a low-finesse Fabry–Pérot cavity. They designed a compact QTF-based FPI module and used a dual-wavelength demodulation method with the ellipse-fitting differential-cross-multiplication algorithm for the FPI measurement. Such an all-optical LITES sensor system improved the SNR by ~3 times, with respect to the conventional electrical LITES sensor system, and achieved an MDL of 422 ppb for H2S detection at 1576.29 nm. Since the QTF has a wide spectral response range and the probe laser of the fiber optic FPI technique is independent of the excitation light for gas absorption, the all-optical LITES sensor system has the potential to be applied to remote and non-contact gas detection in a wide spectral range.

Figure 14. (a) The principle and photograph of the fiber-coupled sensing probe. (b) Experimental setup of the fiber-coupled sensing probe-based LITES sensor system. DFB laser: distributed feedback diode laser; CD: laser current driver; TC: laser temperature controller; DAQ: data acquisition card; LIA: lock-in amplifier; QTF: quartz tuning fork; SMF: single mode fiber. Reproduced with permission from Ref. [135]. Copyright 2021, copyright Elsevier.

In 2022, Pan et al. combined the highly sensitive fiber optic Fabry–Pérot interferometry (FPI) technique with LITES to develop an all-optical LITES sensor system [138], which exploited the FPI method to measure the vibration of the QTF, instead of the conventional piezoelectric detection, as shown in Figure 15. The end surface of the optical fiber and the silver-coated surface of the QTF’s prong formed a low-finesse Fabry–Pérot cavity. They designed a compact QTF-based FPI module and used a dual-wavelength demodulation method with the ellipse-fitting differential-cross-multiplication algorithm for the FPI measurement. Such an all-optical LITES sensor system improved the SNR by ~3 times, with respect to the conventional electrical LITES sensor system, and achieved an MDL of 422 ppb for H2S detection at 1576.29 nm. Since the QTF has a wide spectral response range and the probe laser of the fiber optic FPI technique is independent of the excitation light for gas absorption, the all-optical LITES sensor system has the potential to be applied to remote and non-contact gas detection in a wide spectral range.

Figure 15. Experimental setup of the all-optical LITES sensor system based on the FPI technique. FC: fiber-coupled collimator; NV: needle valve; PC: personal computer. Reprinted/adapted with permission from Ref. [135]. Copyright 2022, copyright Elsevier.

8. Conclusions and Discussions

LITES sensors have the merits of high sensitivity, low cost, small size, wide spectral response range, and non-contact measurement ability. With such extensive research, the
LITES technique has made great development. In this review, the research progress of the LITES technique in recent years has been summarized and discussed. To improve the detection sensitivity of the LITES sensor, a QTF with low pressure operation and a multipass absorption cell with a long optical pathlength can be used. Moreover, the detection sensitivity can be further improved by the combination of QEPAS and LITES, the use of multi-QTF, material-coated QTFs, or custom QTFs. On the other hand, 3D printing, optical fiber structure, and multi-gas detection techniques can be employed to promote the practical application of LITES sensor. Table 2 summarizes the detection performance of the six types of LITES sensors in this review.

Table 2. The detection performance of the six types of LITES sensors in this review. atm: atmospheric pressure.

<table>
<thead>
<tr>
<th>Sensor Type</th>
<th>Target Gas</th>
<th>Resonance Frequency of QTF (kHz)</th>
<th>Q-Factor</th>
<th>Excitation Wavelength (nm)</th>
<th>Effective Optical Pathlength (m)</th>
<th>Operating Pressure of QTF</th>
<th>MDL (ppm)</th>
<th>NNEA (cm$^{-1}$ W/$\sqrt{Hz}$)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional LITES</td>
<td>C$_2$H$_2$</td>
<td>30.71</td>
<td>12,136</td>
<td>1530</td>
<td>0.2</td>
<td>1 atm</td>
<td>0.718 @ 1 s</td>
<td>7.63 × 10$^{-9}$</td>
<td>[66]</td>
</tr>
<tr>
<td></td>
<td>CO</td>
<td>30.71</td>
<td>50,177</td>
<td>2330</td>
<td>10.1</td>
<td>20 Torr</td>
<td>0.47 @ 60 ms</td>
<td>7.4 × 10$^{-10}$</td>
<td>[67]</td>
</tr>
<tr>
<td></td>
<td>CH$_4$</td>
<td>32.78</td>
<td>27,205</td>
<td>1653</td>
<td>4.2</td>
<td>8 Torr</td>
<td>0.052 @ 300 ms</td>
<td>2.1 × 10$^{-8}$</td>
<td>[85]</td>
</tr>
<tr>
<td>LITES + QEPAS</td>
<td>H$_2$O</td>
<td>~32</td>
<td>~10,000</td>
<td>1368</td>
<td>1</td>
<td>1 atm</td>
<td>~12</td>
<td>8.4 × 10$^{-7}$</td>
<td>[104]</td>
</tr>
<tr>
<td>Two-QTF-based LITES</td>
<td>C$_2$H$_2$</td>
<td>30.71</td>
<td>11,503</td>
<td>1530</td>
<td>0.2</td>
<td>1 atm</td>
<td>0.97 @ 1 s</td>
<td>/</td>
<td>[112]</td>
</tr>
<tr>
<td>Material-coated QTF-based LITES</td>
<td>CH$_4$</td>
<td>32.73</td>
<td>9417</td>
<td>1650</td>
<td>0.2</td>
<td>1 atm</td>
<td>0.88 @ 1 s</td>
<td>2.2 × 10$^{-10}$</td>
<td>[113]</td>
</tr>
<tr>
<td></td>
<td>CO$_2$</td>
<td>32.74</td>
<td>6823</td>
<td>1580</td>
<td>20</td>
<td>1 atm</td>
<td>600 @ 100 ms</td>
<td>5.78 × 10$^{-10}$</td>
<td>[114]</td>
</tr>
<tr>
<td>Custom QTF-based LITES</td>
<td>C$_2$H$_2$</td>
<td>9.35</td>
<td>9080</td>
<td>1530</td>
<td>0.2</td>
<td>1 atm</td>
<td>~0.325 @ 1 s</td>
<td>9.16 × 10$^{-10}$</td>
<td>[68]</td>
</tr>
<tr>
<td></td>
<td>CO</td>
<td>2.89</td>
<td>5454</td>
<td>4587</td>
<td>~10.13</td>
<td>1 atm</td>
<td>0.75 × 10$^{-3}$ @ 200 ms</td>
<td>/</td>
<td>[128]</td>
</tr>
<tr>
<td>Optical fiber-based LITES</td>
<td>CH$_4$</td>
<td>32.76</td>
<td>10,476</td>
<td>1653</td>
<td>~0.3</td>
<td>1 atm</td>
<td>~48.8 @ 300 ms</td>
<td>9.66 × 10$^{-9}$</td>
<td>[69]</td>
</tr>
<tr>
<td></td>
<td>CH$_4$</td>
<td>32.76</td>
<td>11,374</td>
<td>1653</td>
<td>~0.3</td>
<td>1 atm</td>
<td>~11 @ 300 ms</td>
<td>/</td>
<td>[135]</td>
</tr>
<tr>
<td></td>
<td>H$_2$S</td>
<td>32.78</td>
<td>10,283</td>
<td>1576</td>
<td>~3.3</td>
<td>1 atm</td>
<td>0.42 @ 300 ms</td>
<td>4.6 × 10$^{-10}$</td>
<td>[138]</td>
</tr>
</tbody>
</table>

Among the six types of LITES sensors we summarized, the conventional LITES sensors used the commercial QTF, which has the simple sensor system structure and does not require any modification of the QTF. The QEPAS-LITES sensors and multi-QTF-based LITES sensors can improve the detection sensitivity, but complicate the sensor system. The material-coated QTF can effectively increase the sensitivity and range of the spectral response, but it requires a complex process to coat the QTF with suitable materials. The custom QTFs with novel structural designs showed a higher SNR than commercial standard QTF and can combine the suitable coating to achieve the wider spectral response range, but the custom QTFs have higher costs and lower yields, compared with commercial QTFs, due to the limitations of the current manufacturing process. In fiber-based LITES sensors, the use of optical fiber can allow the sensor realize long-distance in situ gas detection and immune to electromagnetic interference. However, there are still some problems. When the special optical fibers are used as the gas cell, the high mode interference noise needs to be solved. For the fiber-coupled free-space gas cell, it is difficult to combine with the mid-infrared or THz light source, due to the limitations of current optical fiber fabrication process. As the fiber optic FPI technique is used to measure the vibration of the QTF, the FPI detection unit is complex and expensive, compared with the electrical detection method. Therefore, for different application needs, different sensor systems should be chosen.

All-in-all, a lot of research has shown that LITES-based gas sensors have superior performances by using QTF as a photothermal detector, instead of as commercial wavelength-selective photodetectors in direct absorption spectroscopy (DAS). Moreover, LITES over-
comes the disadvantage of another QTF-based technique (i.e., QEPAS), in that the QTF must contact the target gas; thus, LITES can avoid corrosion of the QTF and realize remote gas sensing (e.g., remote gas leakage detection). Due to these advantages of LITES, it has great potential for usability in real-world applications, such as environmental monitoring, combustion diagnostics, petrochemical industry, and so on. However, in order to improve the detection sensitivity, the LITES technique needs to rely on the expensive and bulky multipass adsorption cell, which limits its low-cost and miniaturization sensing applications (e.g., highly sensitive gas detection in confined spaces). The comparison between LITES and QEPAS is shown in Table 3.

Table 3. Comparison between QEPAS and LITES.

<table>
<thead>
<tr>
<th>Technique</th>
<th>Excitation Signal</th>
<th>Multipass Adsorption Cell</th>
<th>Acoustic Micro-Resonator [37]</th>
<th>Target Gas Contacts QTF</th>
<th>Remote Sensing Ability</th>
</tr>
</thead>
<tbody>
<tr>
<td>QEPAS</td>
<td>Acoustic wave</td>
<td>Not required</td>
<td>Required (to improve the detection sensitivity)</td>
<td>Required</td>
<td>No</td>
</tr>
<tr>
<td>LITES</td>
<td>Electromagnetic radiation</td>
<td>Required (to improve the detection sensitivity)</td>
<td>Not required</td>
<td>Not required</td>
<td>Yes</td>
</tr>
</tbody>
</table>

As a detector, the QTF has the merits of low cost and a small-size and can achieve a wide spectral response range without any cooling system. However, the QTF vibration requires the excitation light to be modulated in the frequency response range of the QTF and the vibration amplitude to reach the maximum at the resonance frequency of the QTF, which is one of the reasons why the QTF detector cannot completely replace the commercial photodetector. In fact, for most gas sensing systems, modulation and demodulation techniques are used to improve the detection signal-to-noise ratio. In this case, the excitation light source needs to be modulated (intensity modulation or wavelength modulation). In terms of LITES-based multi-gas detection technique, time-division multiplexing can be used for time-sharing detection, or frequency-division multiplexing can be used for simultaneous detection, without replacing the detector, due to the excellent wide spectrum response characteristics of QTF, which is a superiority to which the photodetector-based TDLAS technique cannot compare. However, it needs to be pointed out that, when the frequency-division multiplexing is used, multiple excitation light sources are required to be exploited, which greatly increases the cost, and it is impossible to ensure that the modulation frequency of each excitation light source corresponds to the resonant frequency of the QTF, which sacrifices the detection sensitivity. The limitation makes it difficult for LITES to be applied to some fields of multi-gas simultaneous detection, such as non-invasive medical diagnosis, based on breath multispecies rapid real-time monitoring.

In the future, in our opinion, the development direction of LITES technique can refer to the following aspects: (1) As material-coated QTFs and custom QTFs have higher detection sensitivities than standard QTFs, and most gas molecules have a strong “finger-print” absorption spectrum in the mid-infrared region, with the increasingly mature products of high-performance mid-infrared lasers, combining high-performance mid-infrared lasers with excellent material-coated/custom QTFs can achieve highly sensitive atmospheric gas remote sensing (e.g., vehicle exhaust remote sensing), which can avoid using multipass absorption cells to improve detection sensitivity. (2) For low-cost and miniaturized gas sensing applications, the miniaturized multipass absorption cell design can provide a good solution, such as the multipass absorption cell in Refs. [85,101]. The use of 3D printing techniques to integrate the excitation light source, gas cell, and QTF into a module can greatly promote the practicality of LITES technique. On the other hand, with the development of the special optical fiber manufacturing technique, longer optical fiber chambers (e.g., hollow-core fiber) can be employed to improve detection sensitivity and
reduce the sensor size. (3) Since the QTF can respond to THz light, with the development of THz technology, the THz light source could be exploited in the LITES sensor as an excitation light source, which can realize gas sensing in smoke and dust environments, due to the strong penetration of THz light. (4) QTF photothermal detectors, with a wide spectral response range, can be easily integrated in a fast Fourier transform spectrometer, which can avoid the change of detector when varying the spectral range of investigation. (5) The small size and the wide spectral response range of the QTF can provide a promising approach to realize a portable broadband spectrometer for multiple species analysis, which can be applied in environmental monitoring, breath analysis, industrial process control, and other fields.

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