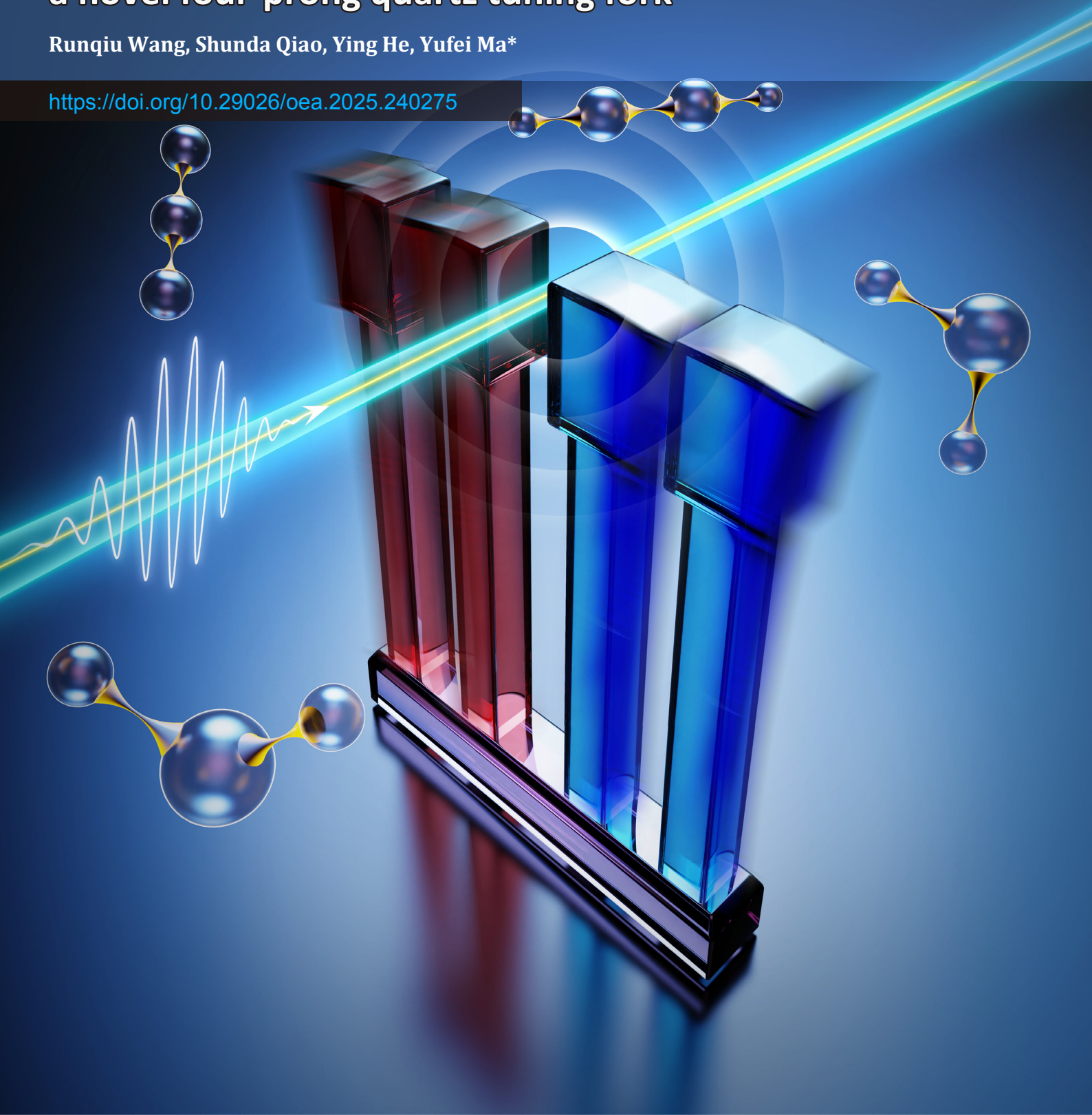


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Highly sensitive laser spectroscopy sensing based on a novel four-prong quartz tuning fork

Runqiu Wang^{1,2}, Shunda Qiao^{1,2}, Ying He^{1,2} and Yufei Ma^{1,2*}

In this paper, a novel four-prong quartz tuning fork (QTF) was designed with enlarged deformation area, large prong gap, and low resonant frequency to improve its performance in laser spectroscopy sensing. A theoretical simulation model was established to optimize the design of the QTF structure. In the simulation of quartz-enhanced photoacoustic spectroscopy (QEPAS) technology, the maximum stress and the surface charge density of the four-prong QTF demonstrated increases of 11.1-fold and 15.9-fold, respectively, compared to that of the standard two-prong QTF. In the simulation of light-induced thermoelastic spectroscopy (LITES) technology, the surface temperature difference of the four-prong QTF was found to be 11.4 times greater than that of the standard QTF. Experimental results indicated that the C₂H₂-QEPAS system based on this innovative design improved the signal-to-noise-ratio (SNR) by 4.67 times compared with the standard QTF-based system, and the SNR could increase up to 147.72 times when the four-prong QTF was equipped with its optimal acoustic micro-resonator (AmR). When the average time of the system reached 370 s, the system achieved a MDL as low as 21 ppb. The four-prong QTF-based C₂H₂-LITES system exhibited a SNR improvement by a factor of 4.52, and a MDL of 96 ppb was obtained when the average time of the system reached 100 s. The theoretical and experimental results effectively demonstrated the superiority of the four-prong QTF in the field of laser spectroscopy sensing.

Keywords: four-prong quartz tuning fork; C₂H₂ detection; quartz-enhanced photoacoustic spectroscopy; light-induced thermoelastic spectroscopy

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Introduction

Highly sensitive detection of trace gas has extensive applications across various fields, including environmental monitoring^{1–5}, medical diagnostics^{6–10}, space exploration^{11–13}, and petrochemical industry^{14–17}. C₂H₂ is an important chemical feedstock as well as a flammable and explosive gas, presenting a significant risk in fire-prone environments. Highly sensitive detection of C₂H₂ can help prevent fire and explosion incidents caused by leaks, thereby protecting both people and equipment. Spectroscopy-based gas sensing technology is an ad-

vanced method for trace gas detection, based on the "fingerprint" absorption spectrum characteristic of gas molecules^{18–22}. It has the advantages of high selectivity, high sensitivity and real-time detection^{23–25}. As one of the spectroscopy-based gas sensing technologies, photoacoustic spectroscopy (PAS) offers a broad linear dynamic range along with nondestructive detection^{26–32}. However, the acoustic cavity used in PAS has a large response bandwidth, making it very sensitive to environmental acoustic noise. In 2002, Tittel et al. proposed quartz-enhanced photoacoustic spectroscopy (QEPAS)³³, utilizing a quartz tuning fork (QTF) as a substitute for the

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traditional microphone to detect sound waves. QEPAS effectively mitigates the restrictions imposed by the acoustic resonance conditions on the structure and size of the gas chamber^{34–37}. The advantages of QEPAS technology lie in its wavelength independence, robustness against external noise, and enabling the analysis of trace gas samples in minuscule volumes^{38,39}. However, due to its contact-based measurement principle, QEPAS is not suitable for detecting corrosive or oxidizing gases, such as hydrogen chloride (HCl) and sulfur dioxide (SO₂). In 2018, Ma et al. put forward a non-contact trace gas detection method called light-induced thermoelastic spectroscopy (LITES)⁴⁰. Since the QTF can be isolated from the target gas, LITES is not only suitable for detecting corrosive gases but also finds applications in remote trace gas detection and combustion diagnostics^{41–45}. QEPAS and LITES are complementary techniques, each suited to different application scenarios^{46,47}.

As the standard QTF was primarily designed for time reference rather than spectral applications, three characteristics of the standard QTF limit its effectiveness in QEPAS and LITES sensors. Firstly, the high resonant frequency (f_0) of 32.768 kHz adversely influences the effective generation of photoacoustic signal while detecting molecules with a slow Vibration-Translation relaxation^{48,49}. Secondly, the high f_0 also results in a relatively short energy accumulation time of the sensor, leading to a reduced amplitude of the system signal⁵⁰. Thirdly, the narrow prong gap spacing of the standard QTF leads to significant optical noise^{51,52}. Current custom QTFs with different sizes are all characterized by large prong gap and low resonant frequency^{53–56}, having superior detection performances to that of the standard QTF.

For the QTF's bending vibration mode used in QEPAS and LITES techniques, the stress induced by the prong vibration tends to concentrate in the narrow area of the internal side where the prongs and the support part are connected⁵⁷. However, the standard QTF and the custom QTFs mentioned above only have a very limited stress concentrated area as each of them only has two prongs. To compensate for this deficiency, the number of prongs can be increased, which would enlarge the contact area and enhance the stress concentration, resulting in a greater piezoelectric charge and a stronger signal.

In this paper, a novel four-prong QTF characterized by a low resonant frequency, a large prong gap, and an expanded thermal expansion area is reported for the first time. The bifurcation of the two prongs into four in-

creases the area of stress concentration, thereby effectively improving the efficiency of acoustic wave detection and producing a large piezoelectric signal under the same excitation conditions. The four-prong QTF was designed based on the finite element simulation and fabricated on a Z-cut quartz crystal wafer with a cutting angle of 2° along the X-axis by photolithography, wet etching, and coating techniques. Acetylene (C₂H₂) was chosen as the target analyte. A C₂H₂-QEPAS system and a C₂H₂-LITES system were built to verify the sensing performance of the four-prong QTF.

QTF simulation

Finite element analysis software was used to design and optimize the geometry of the four-prong QTF, as well as to compare the performance of both the four-prong QTF and the standard QTF in QEPAS and LITES technologies. The design of the QTF should balance the requirements for lower resonance frequency, larger prong spacing, and higher piezoelectric response. The resonant frequency f_0 of the QTF is related to its prong length l and width w according to Euler-Bernoulli model, as shown in Eq. (1)⁵⁸:

$$f_0 = \frac{1.194^2 \pi w}{8\sqrt{12}l^2} \sqrt{\frac{E}{\rho}}, \quad (1)$$

where E and ρ represent the elastic Young modulus and density of quartz, respectively.

In QEPAS, a linear acoustic source was employed to simulate the sound wave generated by the periodic expansion of gas which was heated by a periodically modulated laser. This line acoustic source was placed in the middle of the 2nd and 3rd prongs, 1.7 mm below the prong tip. The physical fields for the simulation of QEPAS mainly contained solid-state mechanics, electrostatic physics and pressure acoustics (frequency domain), and multiple physical fields including acoustic-structure boundary and piezoelectricity. The volumetric flow rate per unit length flowing out from the line acoustic source was set as $i \cdot 8.5 \times 10^{-9} \text{ m}^2/\text{s}$. To intensify the mechanical vibration of the prongs, the frequency of the acoustic source should be consist with the f_0 of the QTF. The first step was to get a succession of different vibration modes in eigenfrequency study. For the standard two-prong QTF, the study focused on the symmetric in-plane vibration mode, as only this mode can generate effective piezoelectric current signals due to its dipole structure. The f_0 of the standard QTF was ascertained as 32633.5 Hz.

For the four-prong QTF, the electrode was designed based on the vibration mode that exhibits the maximum surface charge, optimizing the piezoelectric signal. The four-prong QTF has three in-plane vibration modes: the 1st and 2nd prongs vibrating in the opposite direction to the 3rd and 4th prongs (at 8794.1 Hz) as shown in Fig. 1(a); all four prongs vibrating in the same direction (at 8811.5 Hz) as shown in Fig. 1(b); and the 1st and 2nd prongs vibrating in the opposite direction while the 3rd and 4th prongs also vibrating in the opposite direction with a larger amplitude (at 9048.3 Hz), as shown in Fig. 1(c). The calculated total surface charge of the vibration modes was 5.09×10^{-13} C (at 8794.1 Hz), 4.96×10^{-13} C (at 8811.5 Hz), and 5.94×10^{-14} C (at 9048.3 Hz), respectively. The vibration mode at 8794.1 Hz produced the largest piezoelectric signal, thereby the electrode was configured to operate in this vibration mode for further detection. Subsequently, the frequency domain studies center-

ing around their frequency f_0 were conducted, and the stress distribution of the standard QTF and the four-prong QTF and their dimensions are respectively depicted in Fig. 2(a) and 2(b). For the standard QTF, the stress is concentrated in a very limited zone at the root of the prongs, while for the four-prong QTF, two more prong gaps result in an expansion of the stress concentration area. Meanwhile, each prong has a wider T-head to lower the f_0 and amplify the vibration amplitude. The maximum stress of the four-prong QTF is over one order (11.1 times) of magnitude greater than that of the standard QTF. Moreover, the surface charge density of the four-prong QTF shows a 15.9-fold improvement compared to that of the standard QTF, as depicted in Fig. 3. The total surface charge of the standard QTF and the four-prong QTF are 5.09×10^{-13} C and 1.84×10^{-14} C, respectively, displaying an increase of 27.7 times.

In LITES, the light energy absorbed by the QTF is

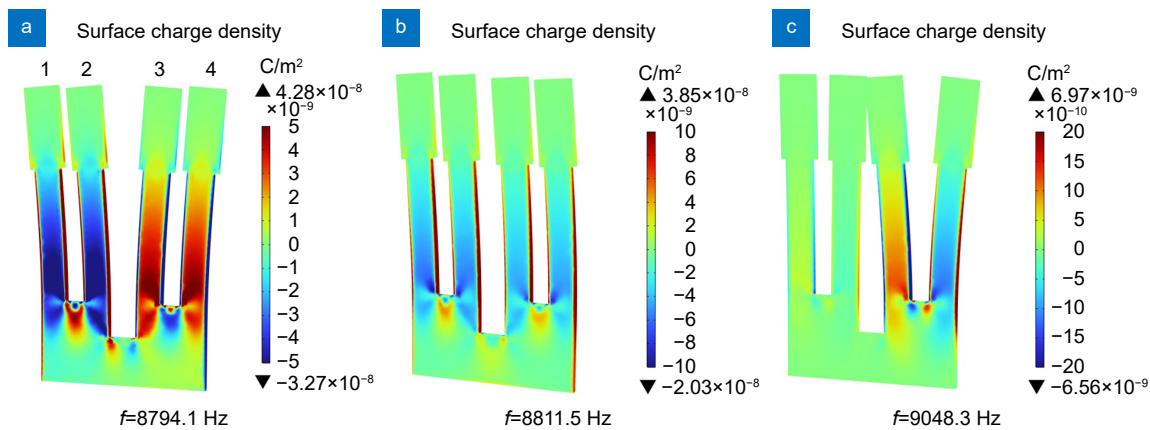


Fig. 1 | Three in-plane vibration modes of the four-prong QTF.

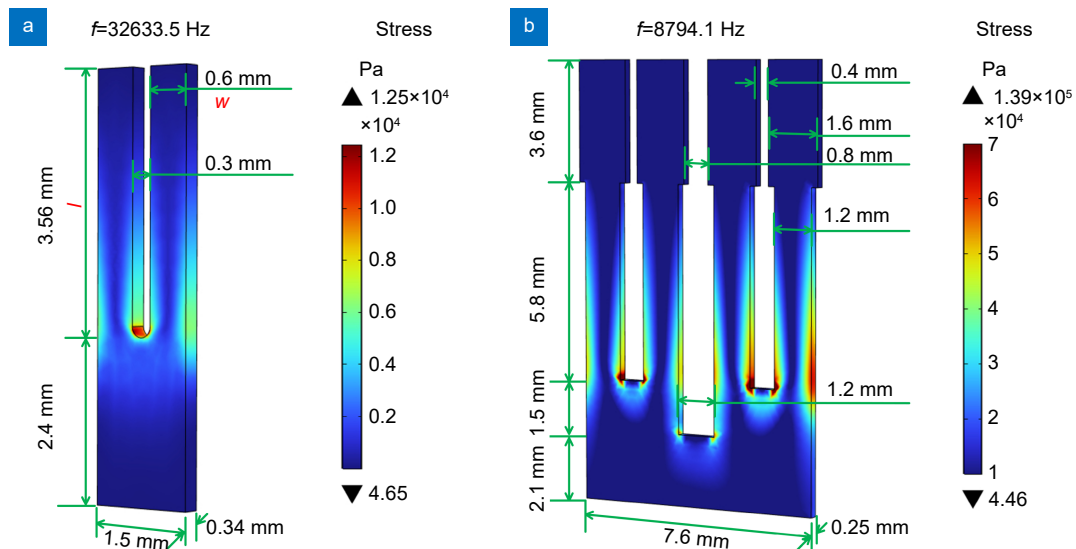


Fig. 2 | The simulation of stress distribution in QEPAS: (a) standard QTF, (b) four-prong QTF.

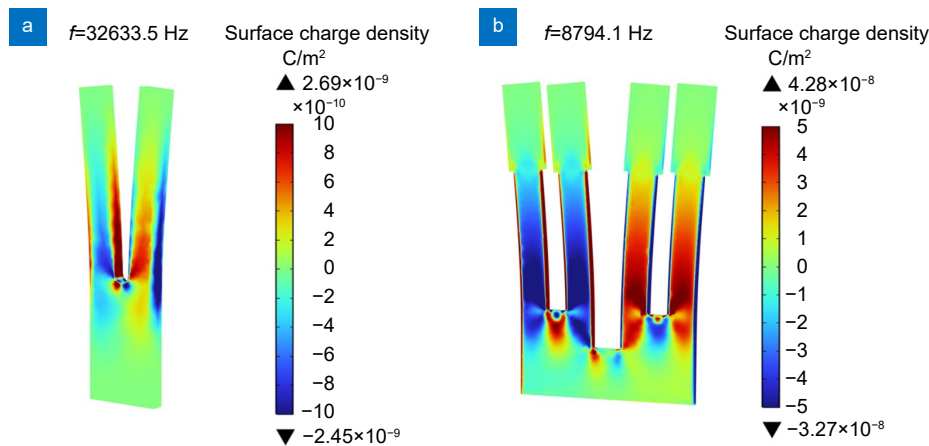


Fig. 3 | The simulation of surface charge density distribution in QEPAS: (a) standard QTF, (b) four-prong QTF.

converted into heat energy, and local thermal expansion in the quartz results in thermoelastic deformation and vibration. Therefore, the simulation for LITES mainly involved physical fields of solid-state mechanics, electrostatic physics, and heat transfer in solids, as well as multi-physical fields including thermal expansion and piezoelectricity. The maximum laser power was set as 5 mW. The whole QTF model was set with charge conservation and grounding conditions. Under the same incident light power, Fig. 4 presents the comparison of temperature distribution between the standard QTF and the four-prong QTF. The surface temperature difference is 7.95×10^{-7} °C for the standard QTF and 9.04×10^{-6} °C for the four-prong QTF, which is 11.4 times greater than that of the former. Fig. 5 illustrates the surface charge density distribution of these two types of QTF, with the peak value of the surface charge density of the four-prong QTF being 11.0 times larger than that of the standard QTF. And the total surface charges of the standard QTF and the four-prong QTF are calculated to be 6.8×10^{-20} C and 1.79×10^{-18} C, respectively, with the in-

crease of 26.32 times for the four-prong QTF. Moreover, the 4-prong QTF had a lower modulation frequency and underwent a longer energy accumulation time when it was exposed to laser irradiation, leading to an increased temperature gradient and a superior detecting performance.

Experimental setup

In this research, a single-mode distributed feedback (DFB) diode laser with an output wavelength of 1530.37 nm was used as the light source to detect a C_2H_2 absorption line located at 6534.36 cm^{-1} . The temperature and the center current of the laser controller were set to 28 °C and 98 mA, respectively. The output power of the laser emitting at 1530.37 nm was measured as 20 mW. The diagrammatic sketch of the C_2H_2 -QEPAS system based on the four-prong QTF is depicted in Fig. 6(a). The wavelength modulation spectroscopy (WMS) and the second harmonic (2f) demodulation technique were employed to reduce the background noise. A low-frequency ramp wave produced by a signal generator and a high-frequency

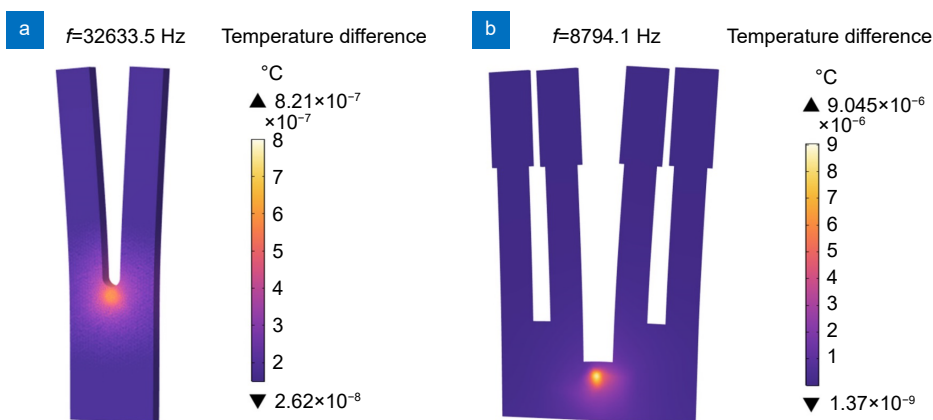


Fig. 4 | The simulation of temperature distribution in LITES: (a) standard QTF, (b) four-prong QTF.

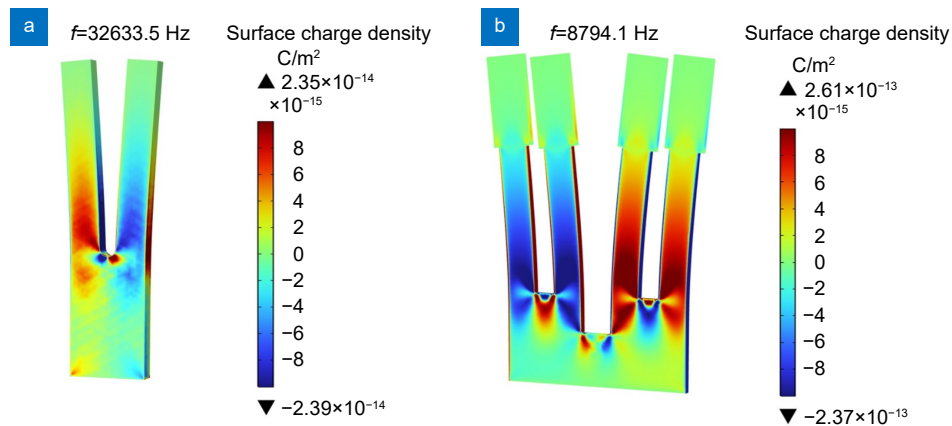


Fig. 5 | The simulation of surface charge density distribution in LITES: (a) standard QTF, (b) four-prong QTF.

sinusoidal wave generated by a lock-in amplifier were added to modulate the output wavelength of the DFB laser. The scanning frequency of the DFB laser was 10 mHz, and the modulating frequency of the laser was $f_0/2$. The tail fiber of the DFB laser was connected to a fiber collimator, and the collimated laser beam entered a gas cell filled with the target gas through a wedge window. After passing through the acoustic micro-resonator (AmR) and the gap of the four-prong QTF, the laser beam eventually entered the power meter for real-time monitoring of the laser transmission to perform optical alignment. The schematic diagram of the C_2H_2 -LITES system based on the four-prong QTF is depicted in Fig. 6(b). The collimated laser passed through a gas chamber with a 20 cm path length and focused on the root of the four-prong QTF by using a focusing lens with a focal

length of 50 mm. The piezoelectric signal produced by the QTF was amplified by a transimpedance amplifier and subsequently transmitted to the lock-in amplifier for second harmonic demodulation. During the experiment, the filter roll-off of the lock-in amplifier was set as fourth order with the detection bandwidth of 324.6 mHz for both QEPAS and LITES system.

Results and discussion

Four-prong QTF-based QEPAS sensor performance

The frequency response characteristics of the standard QTF and the four-prong QTF were investigated by the optical excitation method, as shown in Fig. 7. In QEPAS, the laser beam passed through the prong gap between the 2nd prong and the 3rd prong to generate an acoustic

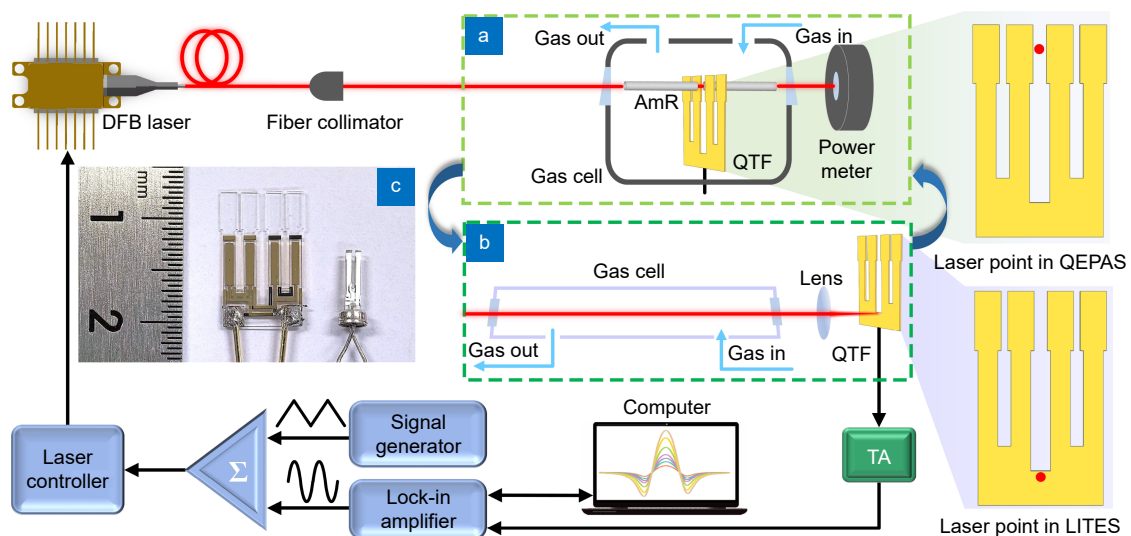


Fig. 6 | Schematic diagram of the C_2H_2 sensor. (a) The sketch of the QEPAS system based on the four-prong QTF. (b) The sketch of the LITES system based on the four-prong QTF. (c) The picture of the four-prong QTF and the standard QTF. AmR: acoustic micro-resonator, TA: transimpedance amplifier, QTF: quartz tuning fork.

wave that propelled the prongs of the QTF. By varying the modulation frequency of the laser to induce acoustic signals from the gas at different frequencies, the frequency response curve of the QTF can be obtained by measuring the piezoelectric signal strength it generates. The frequency at which the curve reaches its maximum value represents the f_0 of the QTF. The measured data were squared, normalized, and then processed by Lorentz fitting in turn, and the frequency corresponding to the peak of the curve represented the fundamental mode frequency of the QTF. The resonant frequency f_0 and bandwidth Δf of the standard QTF were 32767.85 Hz and 3.61 Hz, while those of the four-prong QTF were 7918.98 Hz and 1.02 Hz, respectively. The difference in f_0 between the simulation results and the experimental results can be attributed to the following factors: 1) the f_0 value was influenced by gas damping effects during the experiment; 2) the simulation assumed the QTF was made of quartz, without considering the impact of the gold electrode on the mass distribution of the QTF; 3) the mesh division and constraints applied to the QTF in the simulation affected the f_0 value; 4) a base was welded to the bottom of the QTF to support it and facilitate the export of the piezoelectric signal during the experiments. The quality factor Q , defined by the equation $Q = f_0/\Delta f$, was calculated as 9077 for the standard QTF and 7763 for the four-prong QTF. Compared to the standard QTF, the four-prong QTF exhibits a lower f_0 and Δf , which are advantageous for energy accumulation and noise suppression. Additionally, when the four-prong QTF equipped with AmR, the f_0 had a slight decrease. The modulation depth can influence the detection performance of the sensor. Therefore, it should be optimized. The signal amplitudes of the standard QTF and the four-prong QTF were measured respectively by changing the modulation

current as depicted in Fig. 8. Both signal amplitudes rose first and gradually declined as the modulation current increased. The optimum modulation current for the standard QTF was determined to be 17.58 mA, while for the four-prong QTF, it was 10.67 mA. Usually, the modulation coefficient between the current and wavelength decreases with the frequency. And a high f_0 of the QTF means a high modulation frequency of the laser. Therefore, the optimal modulation current of the standard QTF with a higher f_0 was larger than that of the four-prong QTF.

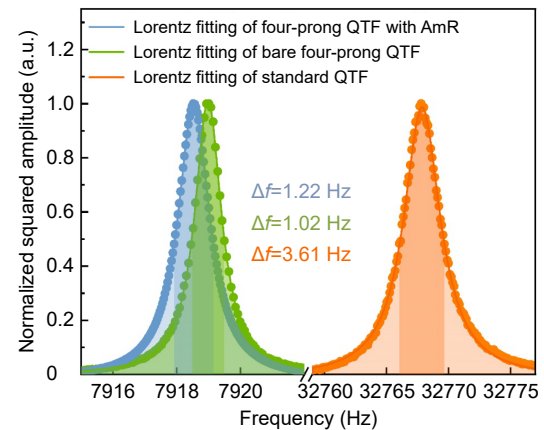


Fig. 7 | The frequency response characteristic of the standard QTF (orange), the bare four-prong QTF (green), and the four-prong QTF equipped with AmR (blue).

To further improve the system performance, the four-prong QTF was configured with a pair of AmR, as shown in Fig. 6(a). The AmR can form an acoustic standing wave cavity, which increases the acoustic wave intensity and enhances the vibration amplitude of the QTF. The optimal length (L) of the resonant tube should be set in the range of $\lambda_s/4 < L < \lambda_s/2$, where λ_s represents the wavelength of the sound wave in the medium⁵⁹. The

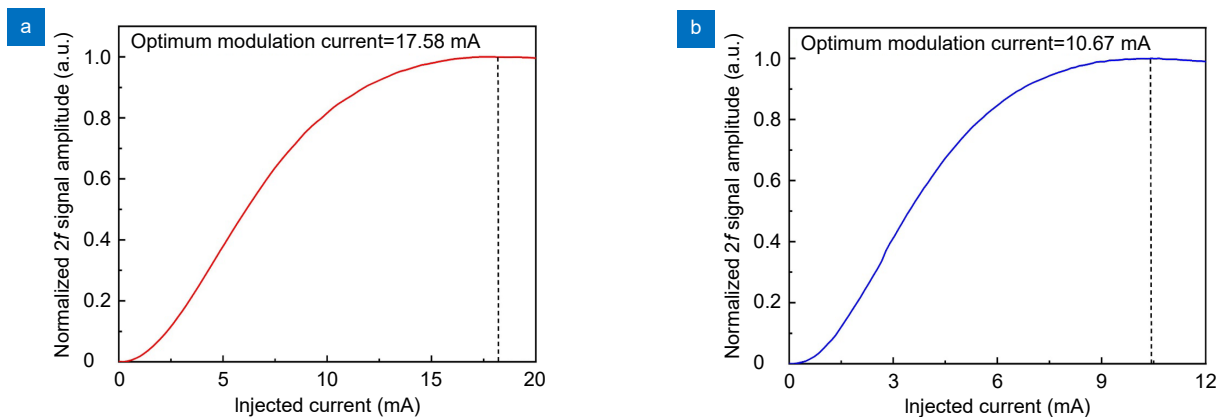


Fig. 8 | The modulation current curves of (a) the standard QTF and (b) the four-prong QTF.

optimization of the tube length and inner diameter of the resonant tube are shown in Fig. 9. The experimental results indicated that the optimal dimensions of the AmR were an inner diameter of 0.9 mm and a length of 20.0 mm. To reduce the energy leakage and improve the optimal acoustic coupling, the distance between the port of the resonator and the QTF should not exceed 50 μm . The f_0 and Δf of the four-prong QTF configured with the optimal AmR were determined as 7918.53 Hz and 1.22 Hz. After being equipped with the AmR, the Q decreased from 7763 to 6490, indicating a strong acoustic coupling between the four-prong QTF and the AmR.

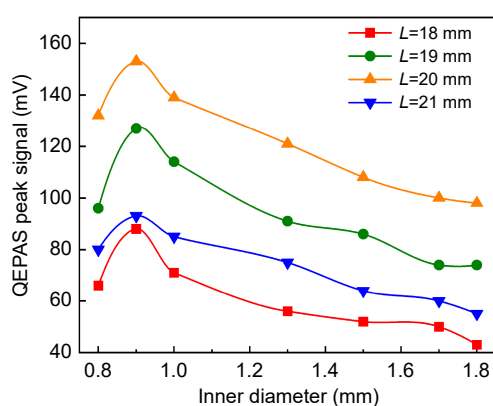


Fig. 9 | The 2f-QEPAS signal peak as a function of the inner diameter and tube length L of the AmR.

The 2f signals of the C_2H_2 -QEPAS system based on the standard QTF, the four-prong QTF, and the four-prong QTF with AmR are shown in Fig. 10(a). The peak values of the 2f signal were 152.67 μV , 740.03 μV , and 153.55 mV respectively, and the signal of the four-prong QTF was 4.85 times higher than that of the standard QTF. This increase could be attributed to the following

reasons. On the one hand, two more prongs brought two more prong gaps where the stress is concentrated, leading to a higher efficiency of acoustic wave excitation. On the other hand, the lower the f_0 of the QTF is, the lower the system modulation frequency becomes, resulting in a longer modulation period. This indicates a longer interaction between the laser and the gas, allowing the target gas to absorb more laser energy and generate a stronger signal. Furthermore, the 2f signal peak of the C_2H_2 -QEPAS system based on the four-prong QTF with AmR was 207.50 times higher than that based on the four-prong QTF. The noise level of the system was measured in a pure nitrogen (N_2) environment as depicted in Fig. 10(b–d). For the C_2H_2 -QEPAS sensors based on the standard QTF, the four-prong QTF, and the four-prong QTF with AmR, the standard deviation (1σ) of the noise were 2.07 μV , 2.15 μV , and 3.02 μV , respectively, and the corresponding SNR were determined to be 73.75, 344.20 and 50844.37, respectively. The minimum detection limits (MDL) can be calculated based on the equation: $\text{MDL} = c/\text{SNR}$, where c is the concentration of the target gas, and SNR is obtained under this concentration. The corresponding MDLs were calculated as 271.18 ppm, 58.11 ppm, and 0.39 ppm, respectively. Compared with the standard QTF-based system, the four-prong QTF-based system improved the SNR by 4.67 times. The SNR of the QEPAS system based on the four-prong QTF with AmR was 147.72 times higher than that of four-prong QTF without AmR.

The 2f signals at different C_2H_2 concentrations were measured, as presented in Fig. 11(a), and the C_2H_2 concentration ranged from 2000 ppm to 20000 ppm. The relationship between the peak value of the 2f signal and the concentration of C_2H_2 is shown in Fig. 11(b), and the R

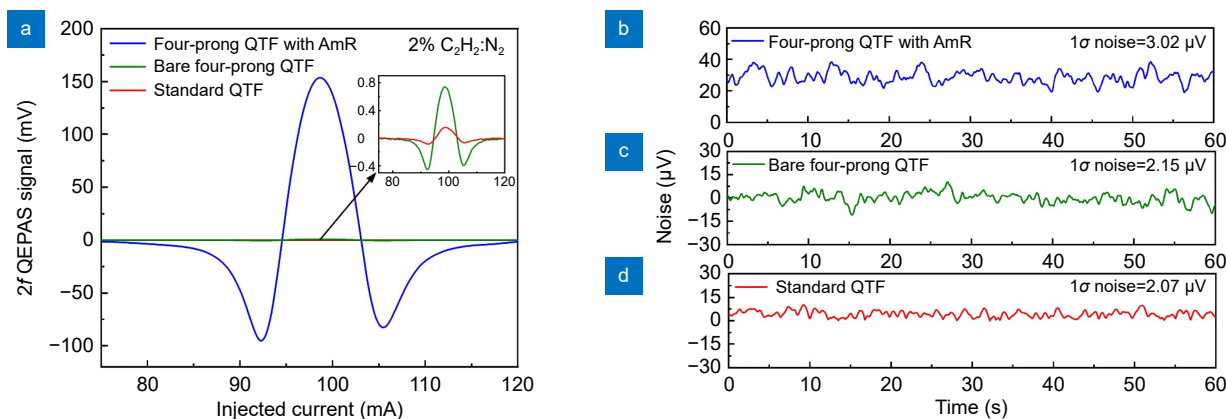


Fig. 10 | (a) The 2f signal of the C_2H_2 -QEPAS system based on the standard QTF, the four-prong QTF, and the four-prong QTF with AmR. (b) The noise level of the C_2H_2 -QEPAS system based on the standard QTF, (c) the four-prong QTF, (d) and the four-prong QTF with AmR.

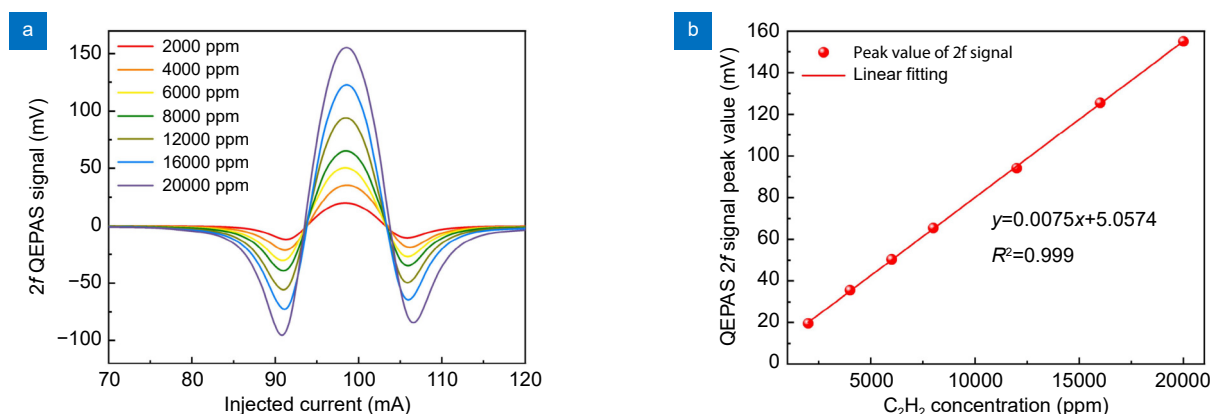


Fig. 11 | The concentration response of the C_2H_2 -QEPAS system based on the four-prong QTF with AmR. (a) 2f signals at different concentrations. (b) The linear relationship between 2f signal peak and C_2H_2 concentration.

square was determined to be 0.999, indicating that the QEPAS sensor based on the four-prong QTF with AmR had an excellent linear concentration response. Finally, to verify the long-term stability of the C_2H_2 -QEPAS sensor of the four-prong QTF with AmR, the gas cell was filled with pure N_2 and tested continuously for 2.5 hours with a time constant of 200 ms. The Allan deviation analysis result is shown in Fig. 12. It can be seen when the average time of the system reached 370 s, the MDL decreased to 21 ppb, indicating excellent detection sensitivity.

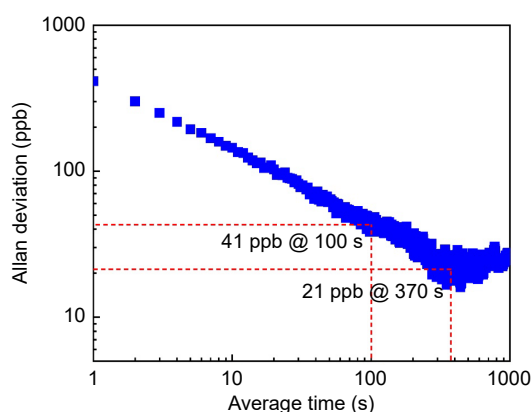


Fig. 12 | The Allan deviation analysis of the C_2H_2 -QEPAS system based on the four-prong QTF with AmR.

Four-prong QTF-based LITES sensor performance

To further verify the detecting performance of the four-prong QTF, the C_2H_2 -LITES system based on the four-prong-QTF was built and is as shown in Fig. 6(b). In LITES, local thermal expansion results in thermoelastic deformation and vibration. The larger geometric dimensions and the enlarged stress accumulation area in the

four-prong QTF are beneficial for enhancing the LITES signal. The 2f-LITES signal at 20000 ppm C_2H_2 concentration was measured at its optimum modulation current, as shown in Fig. 13. The signal amplitude of the standard QTF and the four-prong QTF were 47.34 mV and 247.48 mV, respectively. The signal value of the four-prong QTF was 5.23 times higher than that of the standard QTF. The noise of the system was measured when the gas cell was filled with pure N_2 . For C_2H_2 -LITES sensor based on the standard QTF and the four-prong QTF, the standard deviation of the noise was 4.49 μV and 5.19 μV , respectively. Correspondingly, SNR of 10543.43 and 47684.01 were obtained, and the MDLs were calculated as 1.89 ppm and 0.42 ppm, respectively, for the C_2H_2 -LITES sensor based on the standard QTF and the four-prong QTF. Compared with the LITES system based on the standard QTF, the system based on the four-prong QTF improved the SNR by 4.52 times.

The 2f-LITES signal was measured at different C_2H_2 concentrations, as shown in Fig. 14(a), where the C_2H_2 concentration was ranged from 2000 ppm to 20000 ppm. The relationship between the 2f signal peak and the C_2H_2 concentration is linear, as shown in Fig. 14(b), with an R-square of 0.999, indicating that the LITES sensor based on the four-prong QTF has a perfect linear concentration response. Finally, to validate the stability of the four-prong QTF-based C_2H_2 -LITES sensor, the gas cell was filled with pure N_2 and tested continuously for 2 h with a time constant of 200 ms. The result of the Allan deviation analysis of the system based on the four-prong QTF is shown in Fig. 15. It can be found when the average time of the system reached 100 s, the MDL of the system improved to 96 ppb.

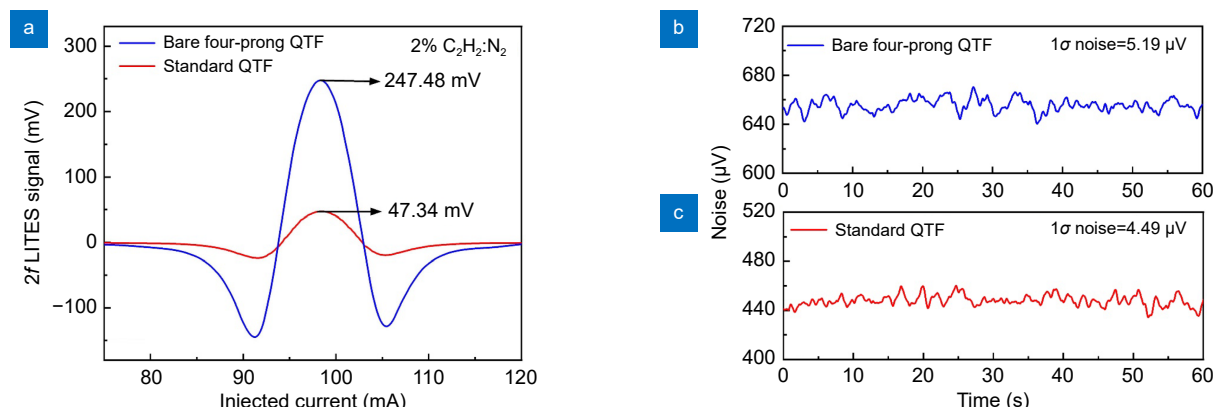


Fig. 13 | (a) 2f signals of the C₂H₂-LITES system based on the standard QTF and the four-prong QTF, the noise level of C₂H₂-LITES system based on (b) the standard QTF, and (c) the four-prong QTF.

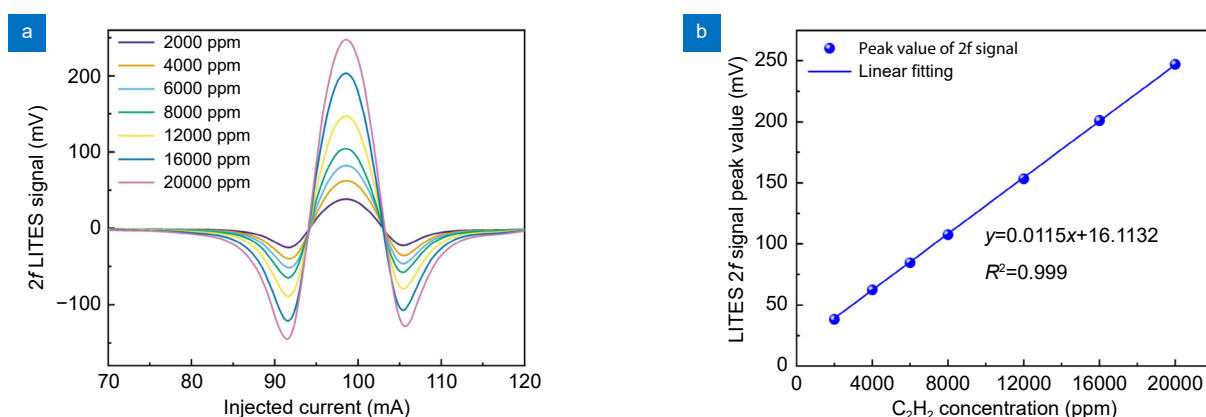


Fig. 14 | The concentration response of the C₂H₂-LITES system based on the four-prong QTF. (a) 2f signals at different concentrations. (b) The linear relationship between 2f signal peak and C₂H₂ concentration.

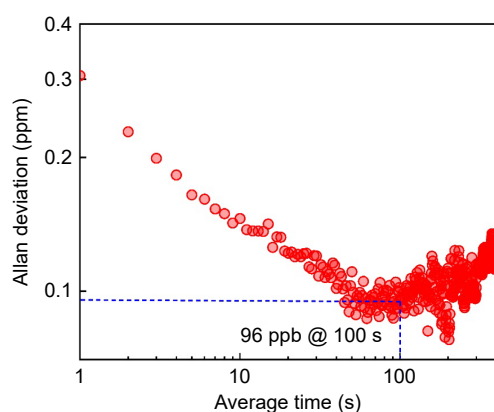


Fig. 15 | The Allan deviation analysis of the C₂H₂-LITES system based on the four-prong QTF.

Conclusions

In conclusion, a novel four-prong QTF was designed and simulated by finite element analysis. By adding the two prongs into four, the area of stress concentration was enhanced, thereby effectively improving the efficiency of

acoustic wave detection and yielding a larger piezoelectric signal under the same excitation conditions. With a low frequency of 7918.98 Hz, a high Q factor of 7763, a large fork prong gap of 0.8 mm, and a T-head for each prong, the energy accumulation time increased and the optical noise was reduced. The simulation results of the QEPAS and LITES technologies indicated that the maximum stress, the maximum surface charge density, and the surface temperature difference of the four-prong QTF were determined to be 11.1 times, 15.9 times, and 11.4 times greater than those of the standard two-prong QTF, respectively. The sensing performance of the C₂H₂-QEPAS system and the C₂H₂-LITES system, employing the standard QTF and the four-prong QTF, were experimentally investigated. For the C₂H₂-QEPAS systems, utilizing the four-prong QTF improved the SNR by 4.67 times compared with the standard QTF-based system, and the SNR could increase up to 147.72 times when the four-prong QTF was equipped with its optimal AmR. When the average time of the system reached 370 s, the

system improved its MDL to 21 ppb. For the C₂H₂-LITES system, the one based on the four-prong QTF exhibited a SNR improvement by a factor of 4.52, and a MDL of 96 ppb was obtained when the average time of the system reached 100 s. The theoretical and experimental results presented in this paper effectively demonstrate the superiority of the four-prong QTF in the field of laser spectroscopy sensing.

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Author contributions

Yufei Ma proposed the original idea, supervised the whole project and revised the manuscript. Runqiu Wang performed the measurements and wrote the original manuscript. Shunda Qiao and Ying He contributed to the discussions.

Competing interests

Yufei Ma serves as an Editor for the Journal, and no other author has reported any competing interests.



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