

Electronic Supplementary Information of

Determination of Broadband-Light Atomic Absorption through Interferometric Spectrometry with a Spatial Heterodyne Spectrometer

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1. Spectrum of the white light background source

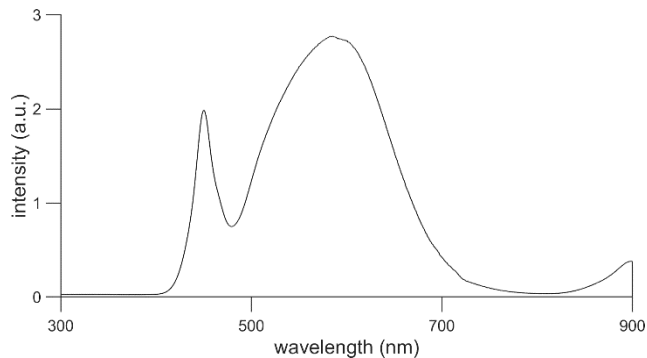


Figure S 1. The spectrum of the white light-emitting diode (LED) used as the broadband background source, recorded by a Czerny-Turner spectrometer (Andor SR-303, Oxford Instrument, Belfast, UK).

2. Interferograms: background versus absorption

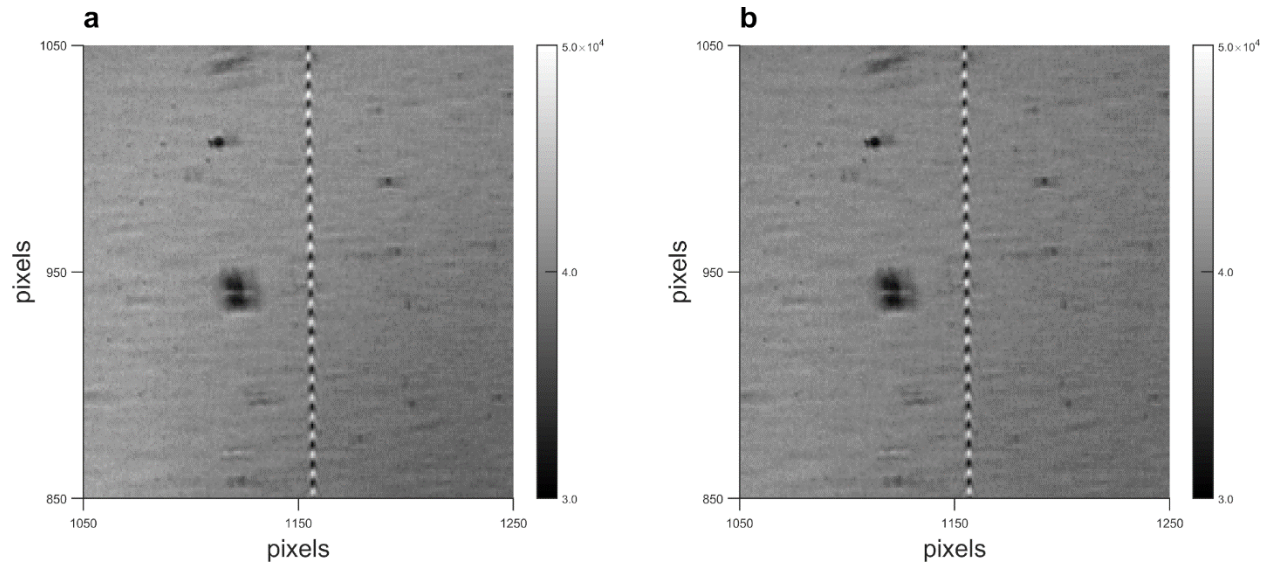


Figure S 2. Background versus absorption under the same zoom condition. a is the raw interferogram of Na D-lines absorption with a white light background field (*cf.* Figure 2a-1). b is the interferogram of the white light background field itself.

3. Interferogram as a function of position and wavelength

The initial equation was of wavenumber σ :

$$I_h(x)|_y = \int_0^{\infty} I(\sigma) (1 + \cos(2\pi(4(\sigma - \sigma_0)\tan\theta \cdot x + 2\alpha\sigma \cdot y))) d\sigma$$

where $I(\sigma)$ is the input spectrum as a function of wavenumber, α is the tilting angle of one of the gratings to ensure a non-zero k_y . θ is the Littrow angle corresponding to σ_0 . To replace σ with wavelength λ , the equation is rewritten into:

$$I_h(x)|_y = \int_0^{\infty} I\left(\frac{1}{\lambda}\right) \left(1 + \cos\left(2\pi\left(4\left(\frac{1}{\lambda} - \frac{1}{\lambda_0}\right)\tan\theta \cdot x + 2\alpha\frac{1}{\lambda} \cdot y\right)\right)\right) d\frac{1}{\lambda}$$

where:

$$\sigma = \frac{1}{\lambda}$$

Then it can be rearranged to:

$$I_h(x)|_y = \int_{\infty}^0 -\frac{1}{\lambda^2} \cdot I\left(\frac{1}{\lambda}\right) \left(1 + \cos\left(2\pi\left(4\frac{\lambda_0 - \lambda}{\lambda_0\lambda}\tan\theta \cdot x + \frac{2\alpha}{\lambda} \cdot y\right)\right)\right) d\lambda$$

Swap the upper and lower bound of the integral, and define the input spectrum as a function of wavelength as $I_s(\lambda)$:

$$I_s(\lambda) = \frac{1}{\lambda^2} \cdot I\left(\frac{1}{\lambda}\right)$$

the final form of interferogram as a function of pixel position and wavelength is written as:

$$I_h(x)|_y = \int_0^{\infty} I_s(\lambda) \left(1 + \cos\left(2\pi\left(4\frac{\lambda_0 - \lambda}{\lambda_0\lambda}\tan\theta \cdot x + \frac{2\alpha}{\lambda} \cdot y\right)\right)\right) d\lambda$$

4. Bandwidth and interferogram

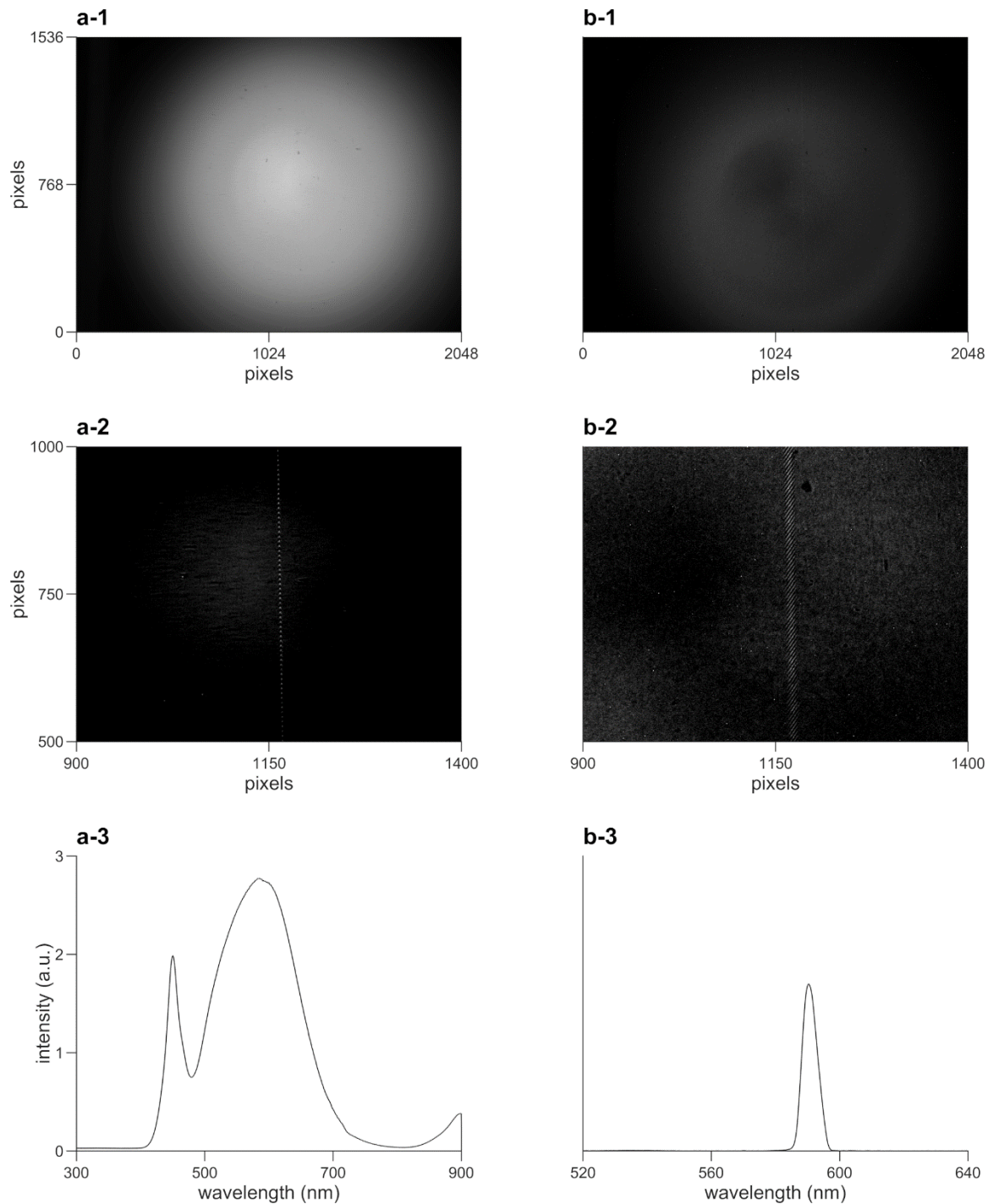


Figure S 3. Interferograms of input fields with different bandwidths. Group a: interferogram and spectrum of the white light background source used in the present study, where a-1 shows the raw image detected by the camera, a-2 is a 500×500 area-of-interest of a-1 and a-3 is the corresponding spectrum recorded by a Czerny-Turner spectrometer (Andor SR-303, Oxford Instrument, Belfast, UK). Group b: interferogram and spectrum of the white light background source through a monochromator, the resulting FWHM is *ca.* 5nm.

5. Periodic variation of the wavelet in the interferogram

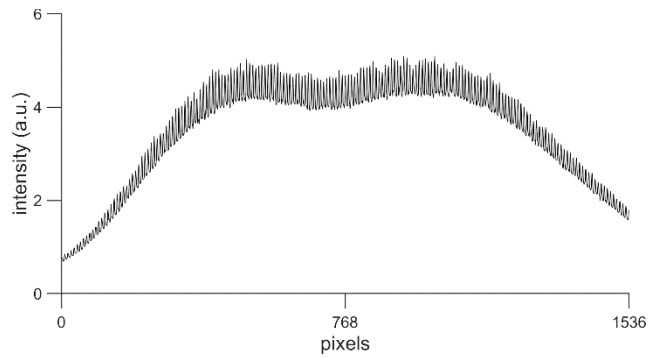


Figure S 4. The section view of the wavelet in the interferogram (*cf.* Figure 2a-1). The periodic variation of the wavelet along the vertical axis results from the on-purpose tilting of one of the gratings, where the period is determined by the tilting angle.

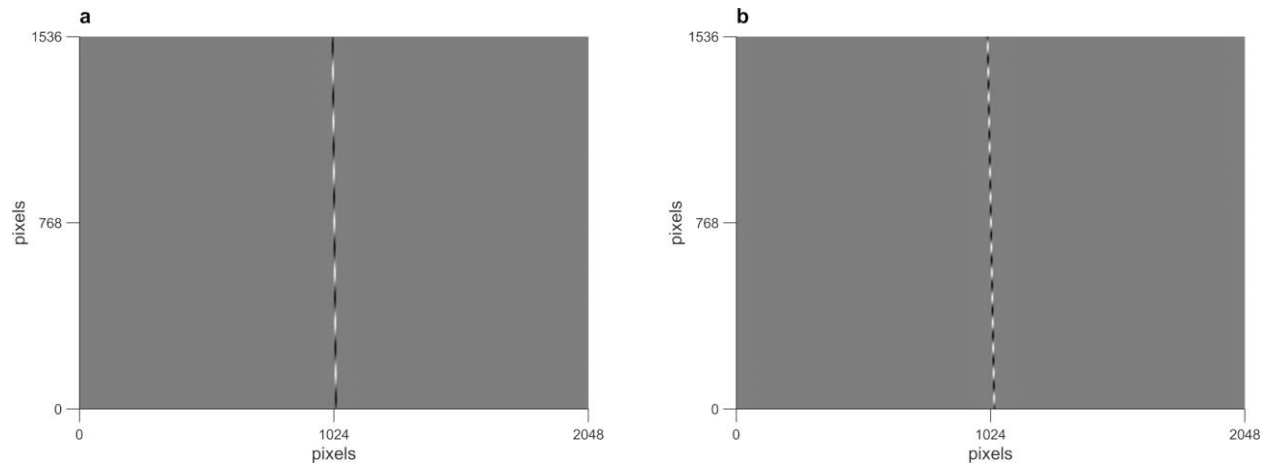


Figure S 5. Simulation of 2D interferogram using Eq. 2. a: tilting angle of the grating, α , is 1° . b: α is 2° . As shown, the period of the only seeable fringes (*i.e.* the wavelet) along the vertical axis is determined by the tilting angle α . The model of input spectrum $I_S(\lambda)$ is set 1 listed in section 6.

6. Models of input spectra used in the simulation

Set 1 Continuous broadband input spectrum with a normalized Gaussian profile (normal distribution), representing a white light input field:

$$I_s(\lambda) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{\lambda-\mu}{\sigma}\right)^2}$$

where $\sigma = 60$ nm, *e.g.* full width half maximum (FWHM) = 141.30 nm, central wavelength $\mu = 579$ nm, same as λ_0 .

Set 2 Similar to Group 1, a continuous input spectrum with normalized Gaussian profile but with narrower bandwidth, where $\sigma = 2$ nm (FWHM = 4.71 nm), central wavelength $\mu = 594$ nm.

Set 3 An idealized Na doublet model with each emission line enveloped by a Gaussian profile normalized to one half:

$$I_s(\lambda) = \frac{1}{2} \cdot \frac{1}{\sigma\sqrt{2\pi}} \left(e^{-\frac{1}{2}\left(\frac{\lambda-\mu_1}{\sigma}\right)^2} + e^{-\frac{1}{2}\left(\frac{\lambda-\mu_2}{\sigma}\right)^2} \right)$$

with $\sigma = 0.1$ nm (FWHM = 0.24 nm), central wavelengths $\mu_1 = 589.0$ nm, $\mu_2 = 589.6$ nm.

Set 4 Normalized broadband input spectrum with Na doublet absorption lines:

$$I_s(\lambda) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{\lambda-\mu}{\sigma}\right)^2} \cdot \left[1 - \frac{1}{2} \cdot \frac{1}{\sigma'\sqrt{2\pi}} \left(e^{-\frac{1}{2}\left(\frac{\lambda-\mu_1}{\sigma'}\right)^2} + e^{-\frac{1}{2}\left(\frac{\lambda-\mu_2}{\sigma'}\right)^2} \right) \right]$$

with $\sigma = 60$ nm, $\mu = 579$ nm; $\sigma' = 0.1$ nm, $\mu_1 = 589.0$ nm, $\mu_2 = 589.6$ nm.

7. Silhouette-based spectral extraction versus column averaging

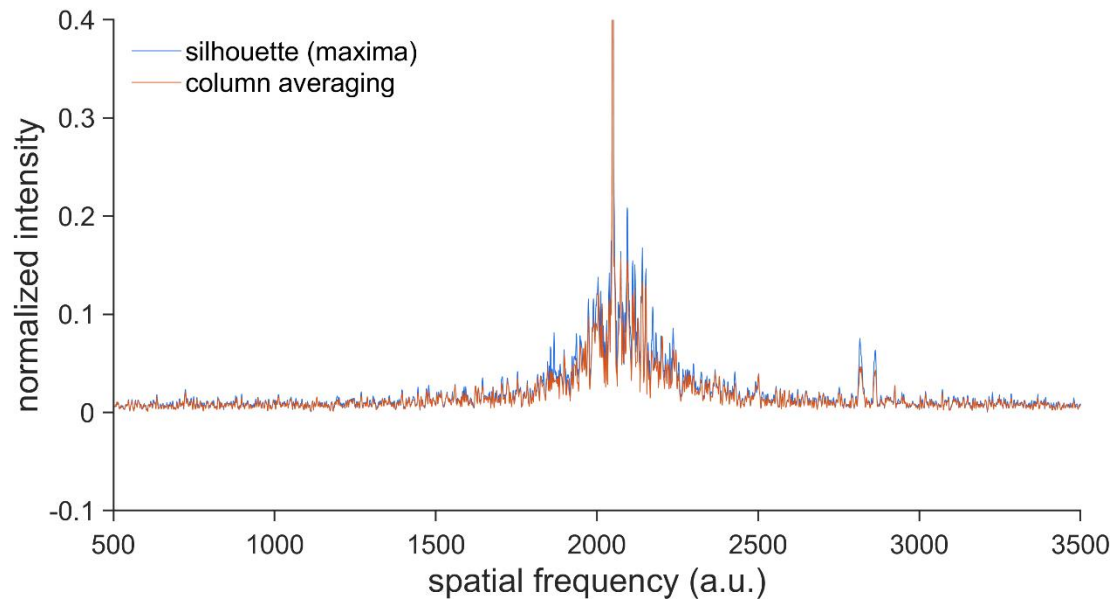


Figure S 6. Spectra extracted from the same interferogram with silhouette and averaging. The spectral-information-rich area (*cf.* Figure 4d, between the white traces) in the Fourier domain is fixed. Spectra were then extracted by finding the maximum or mean value of each column, respectively. The intensities of both extracted spectra are normalized for intuitive comparison. Roughly, the signal-to-background ratio is doubled for silhouette compared to averaging.

8. Pseudocode of broadband-feature detection

The pseudocode for detecting the broadband feature (*cf.* Figure S2) is given in Code S1.

Code S 1.

```
function patch_image is
    input:      uint16[] camera_image
                 double a // slope of the broadband feature
                 double b // intercept of the broadband feature
                 uint16 patch_half_width // manually set to 4

    output:    uint16[] output_image // to be zero-padded and Fourier-
transformed

    constants: image_width
                  image_height

    output_image = camera_image // cloning the image for patching
    double[] y // discrete pixel points representing the image width
    for incremental j from 0 to image_height
        left_index = (j - b) / a - patch_half_width
        right_index = (j - b) / a + patch_half_width
        left_edge_value = camera_image[left_index]
        right_edge_value = camera_image[right_index]
        patching_value = (left_edge_value + right_edge_value) / 2

        for incremental k from left_index to right_index
            uint32 pixel_index = convert_coor_to_index(k, j)
            output_image[pixel_index] = patching_value

function detect_broadband_feature is
    input:      uint16[] camera_image
    output:    double a // slope of the broadband feature
                 double b // intercept of the broadband feature

    constants: num_sampling =150
                  span = 50
                  image_height
                  image_width

    // calculate horizontal
    camera_image_differential = horizontal_differential_abs(camera_image)

    double[] X, Y
    for incremental j from 0 to num_sampling
```

```
index = random integer between 0 and image_height
image_section = full-width image in the region from (index - span) to
                (index + span)
v_projection = vertically binned image_section
v_projection_smooth = boxcar-smoothed v_projection
peak_coor = the x-coordinate where the maximum of
            v_projection_smooth occurs
X[j] = index
Y[j] = peak_coor
```

a, b = solve for linear regression Y vs X

function gradient_abs is

input: **uint16[]** image_input

output: **uint16[]** image_output

for incremental i from 0 to image_height

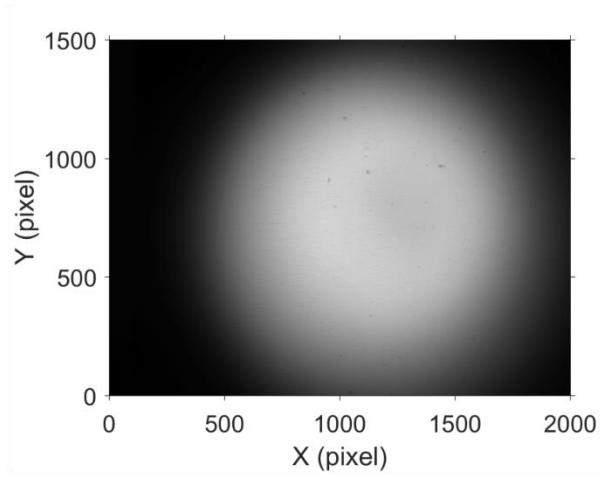
for incremental j from 1 to image_width

 t = i * image_height + j

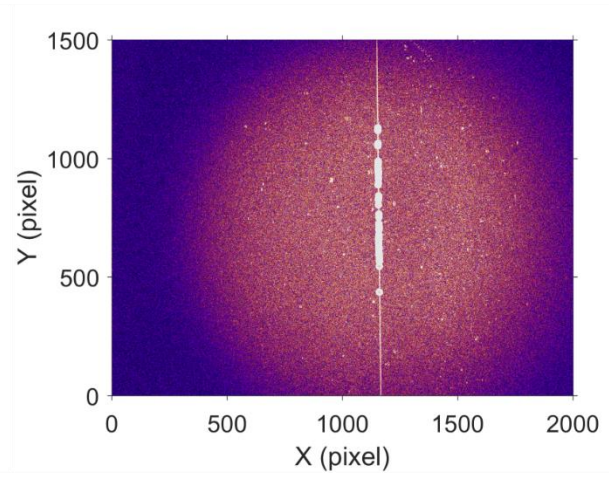
 image_output[t] = abs(image_input[t-1] - image_input[t]);

In the present work, the detection of the broadband feature is semi-automatic. In specific, the slope and intercept of the lines at which the broadband light dominated was found programmatically (*cf.* Figure S7ab). Based on the calculated slope and intercept, each horizontal line the absolute first-order derivative of the raw image can be overlaid (*cf.* Figure S7c). Certainly, the patching width can be determined programmatically, which can be quite useful in many application scenarios. However, in the present study, the patch width can be easily determined upon inspection. In the present study, we manually chose the edges of the broadband features and set it to be 4 px throughout this work.

a) camera_image



b) camera_image_differential (abs)



c) overlay of camera_image_differential

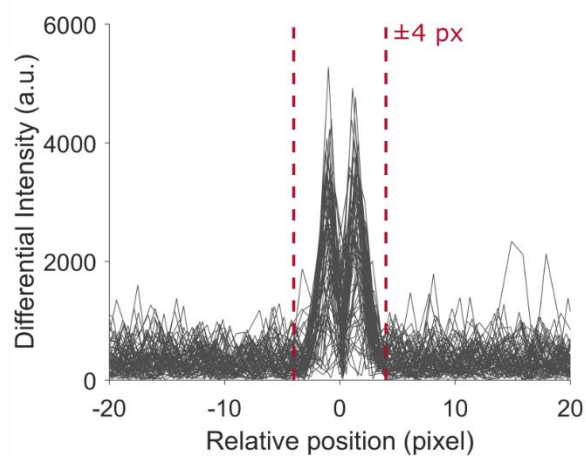


Figure S 7. Function inputs and outputs for automatic broadband-feature-patching.

9. Quantitative assessment

An additional FAA setup was used to determine LoD with single-point calibration. The only difference between this additional setup and the main setup used in the main text was that the quartz cuvette was replaced by a quartz tube (*cf.* Figure S 7). 5 uL of the prepared NaCl solution with a concentration of 1 M was injected into the quartz tube with a 10.00 uL pipette, and was then heated with the propane/butane burner. The interferogram was recorded continuously for an adequate time period to ensure complete consumption of the sample. The exposure time for each frame was 94 ms. For data processing, continuous frames with absorption spectra were selected. The single-point intensity (counts) at 588.9951 nm was integrated over the period of sample introduction. Then mean value of the integrated intensity over time (cps) was used as the calibration point. The corresponding standard deviation was then calculated and shown as the error bar. The calibration line is a straight line through the origin and the calibration point. (*cf.* Figure S 8) To further determine LoD with the calibration above, 3σ of the noise floor was used to represent the signal-to-noise ratio of 3, which corresponded to 9.58 nmol.



Figure S 8. Image of the quartz tube used in the additional FAA setup. The illumination light was fed through a 600- μm diameter optical fiber and a collimator (F810SMA-543, Thorlabs). The collection optics was placed at the downstream of the optical path after the quartz sampling chamber. The beam was then refocused at the collector with a plano-convex lens ($f = 50 \text{ mm}$ LA1131, Thorlabs). The collected light was directed to the spectrometer through another 600- μm optical fiber.

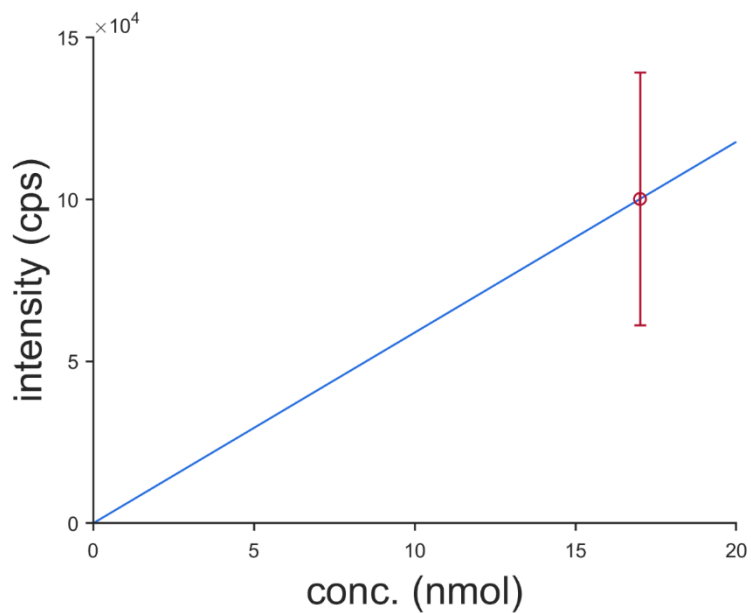


Figure S 9. Determination of the LoD with single-point calibration. The red circle with errorbar is the calibration point and the blue trace is the calibration line, which is a straight line through the origin. The abbreviation “cps” and “conc.” stand for counts-per-second and concentration, respectively.