

Supplementary Information for

**Direction-Resolved Nanoscale Optical Imaging with Near-Nanometer Resolution by
Emerging Infrared Torsional Force Microscopy**

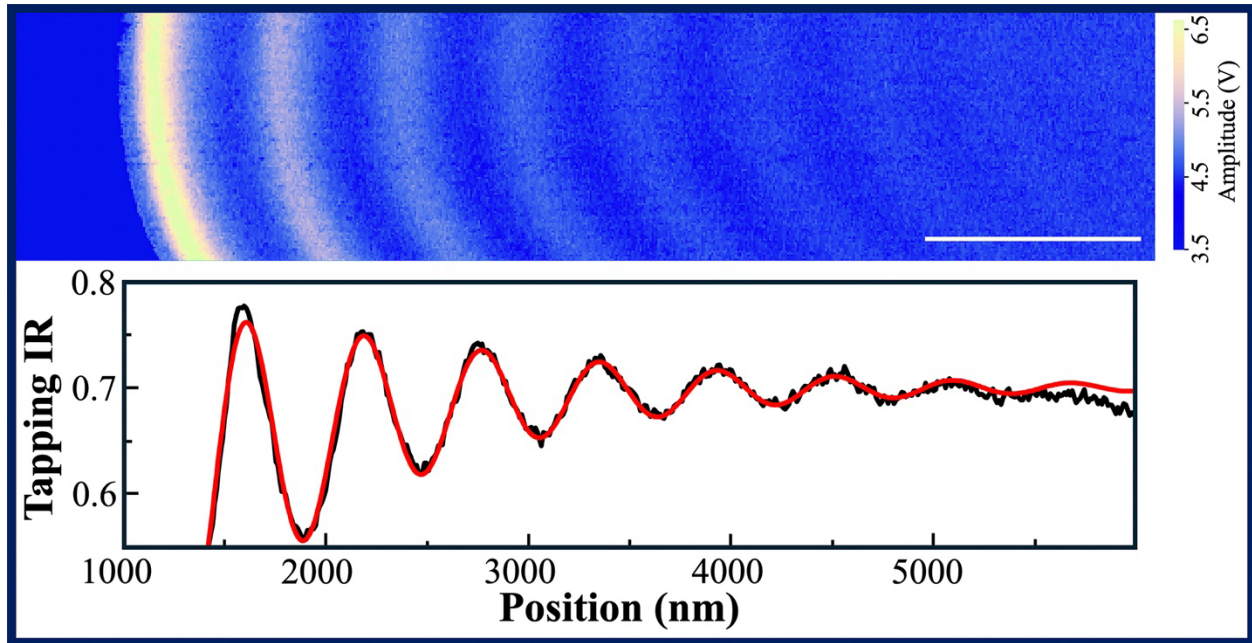
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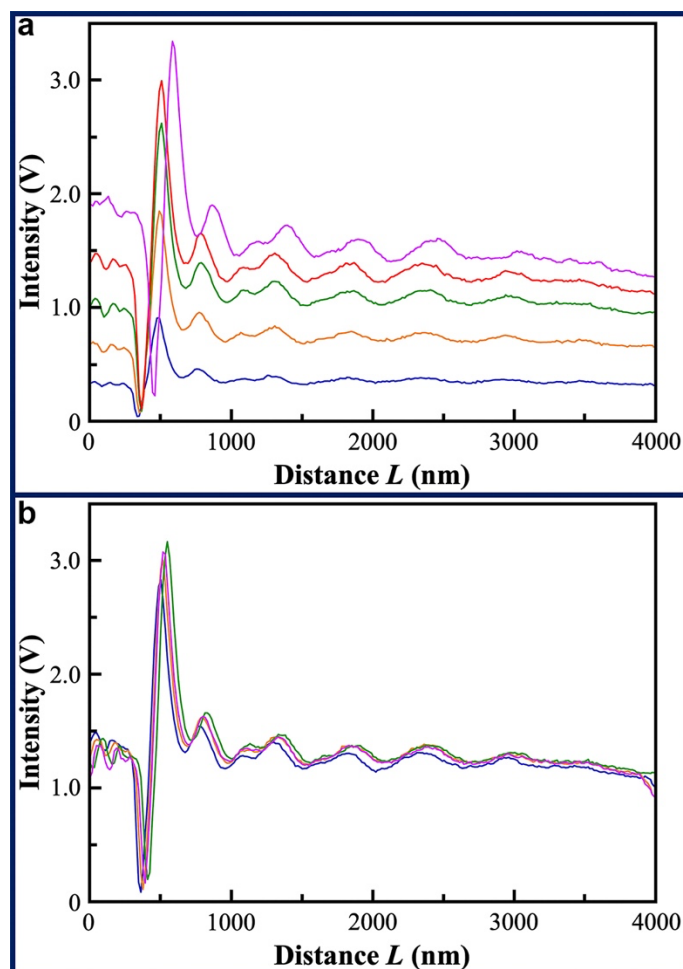
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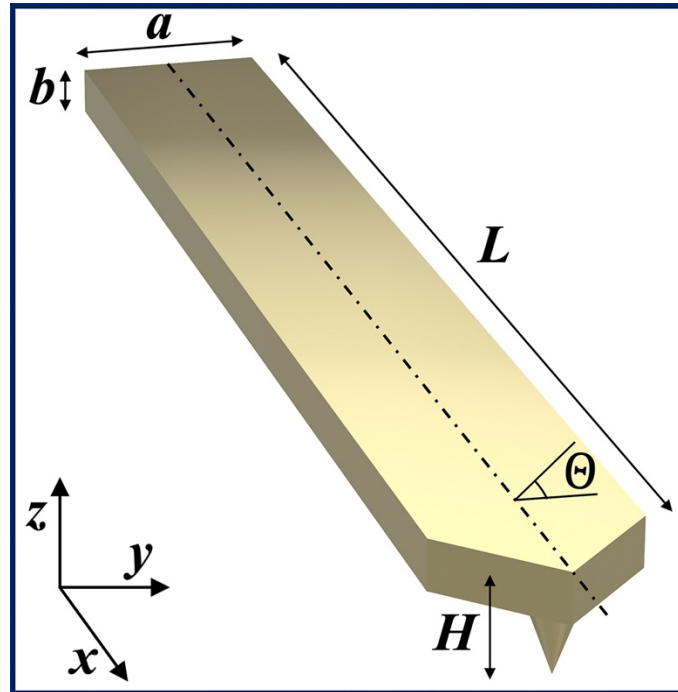
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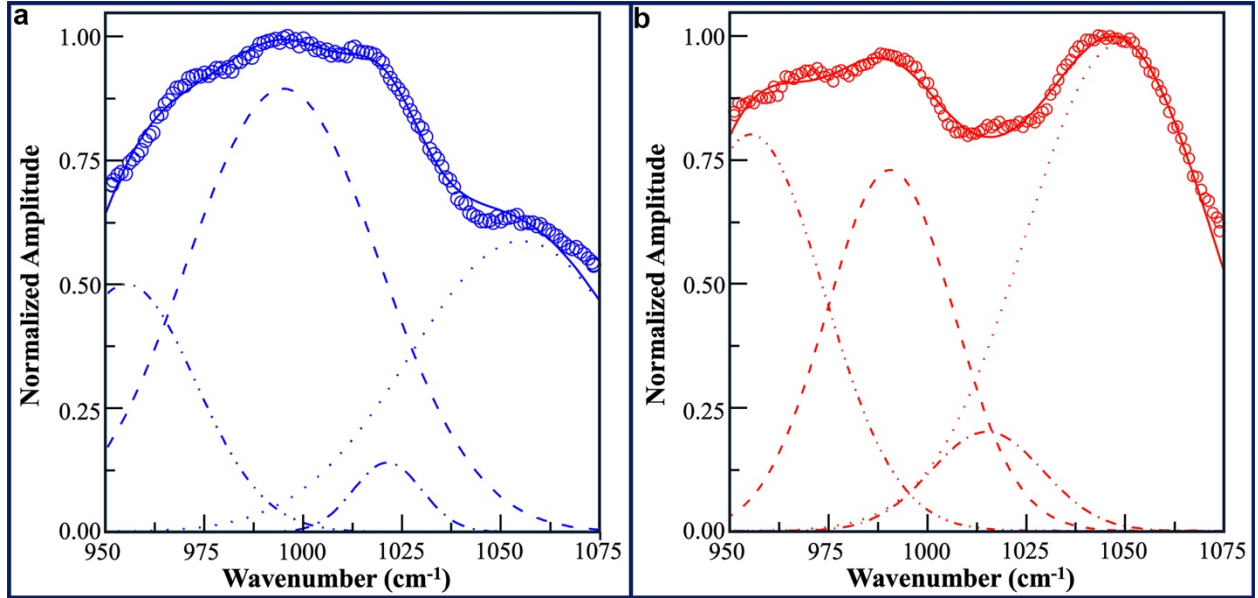
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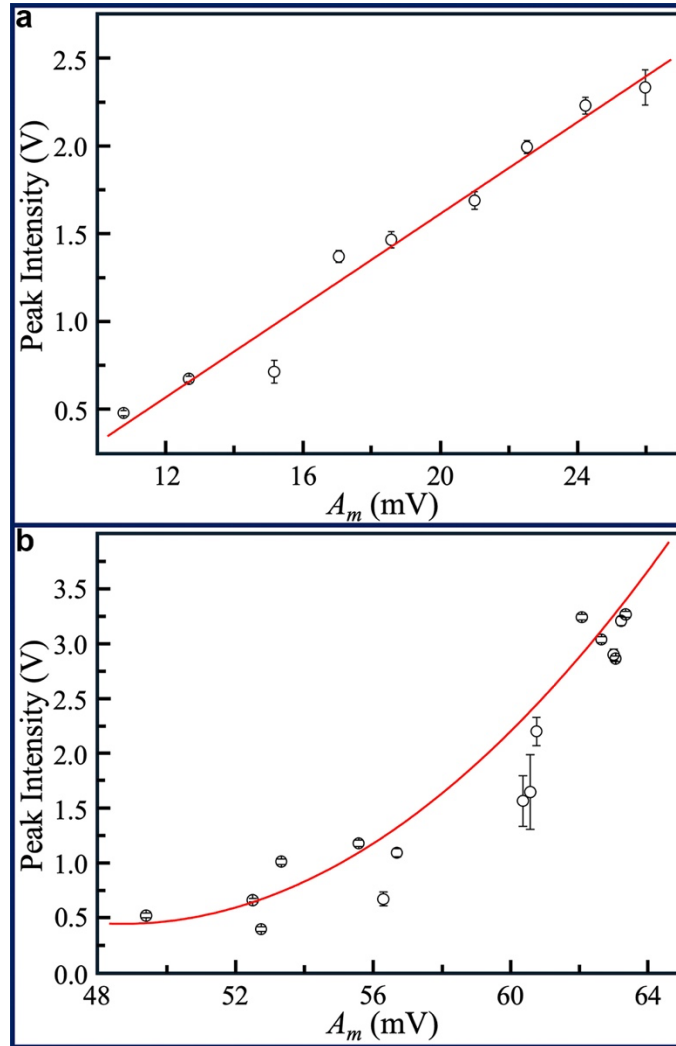
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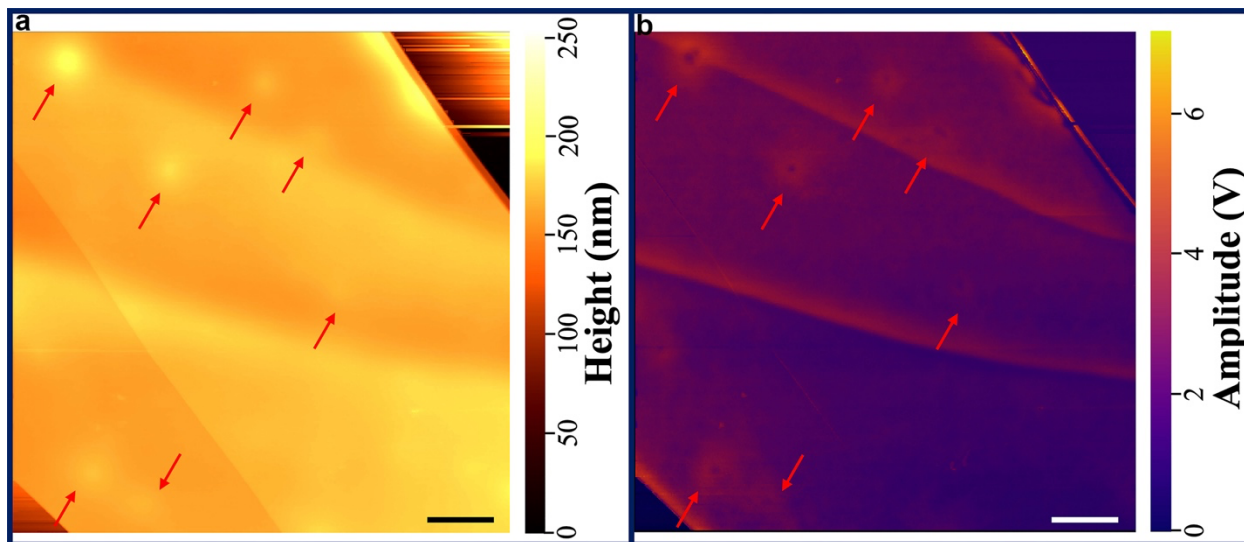
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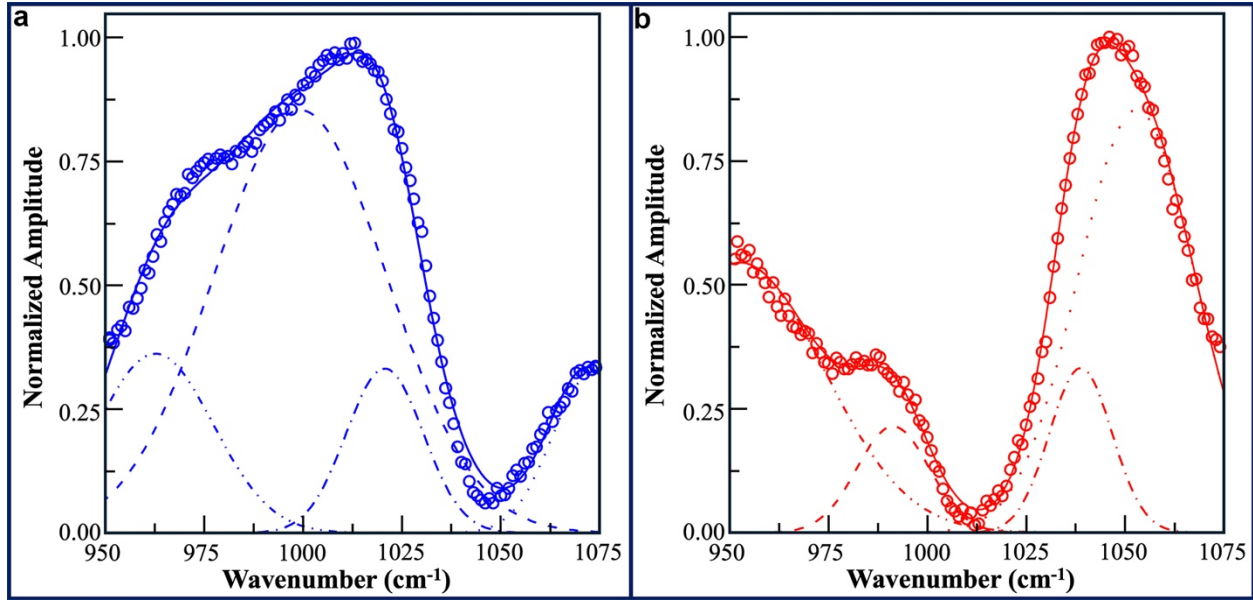
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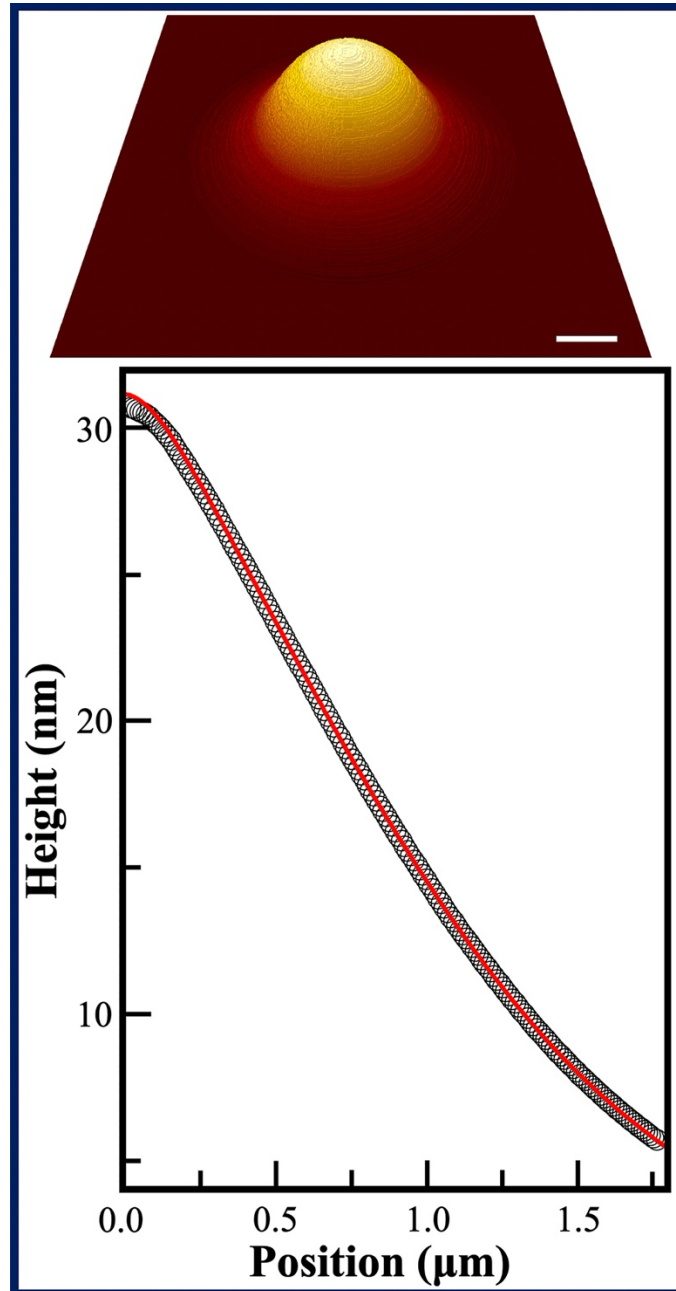
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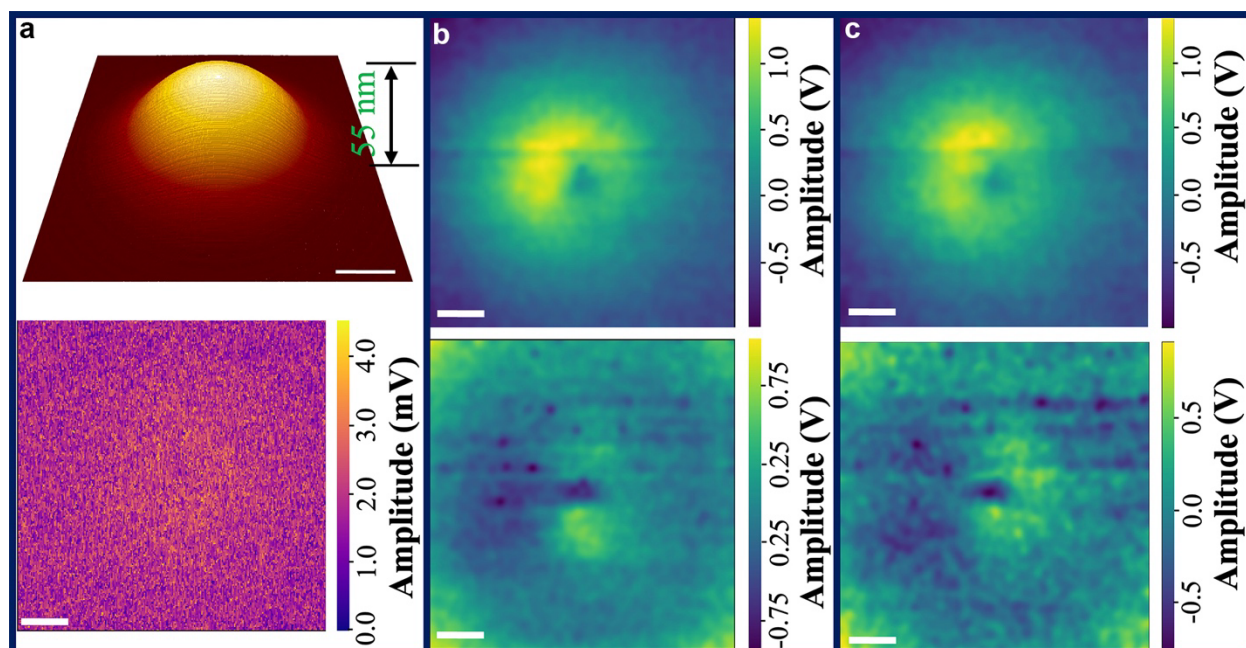
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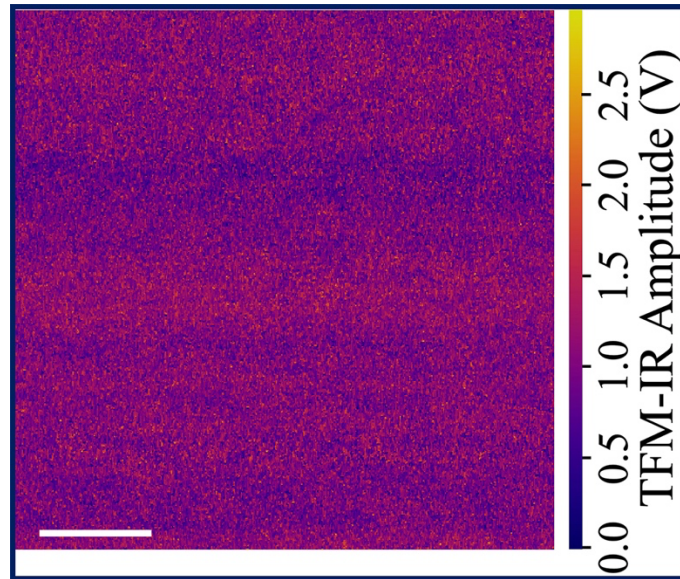
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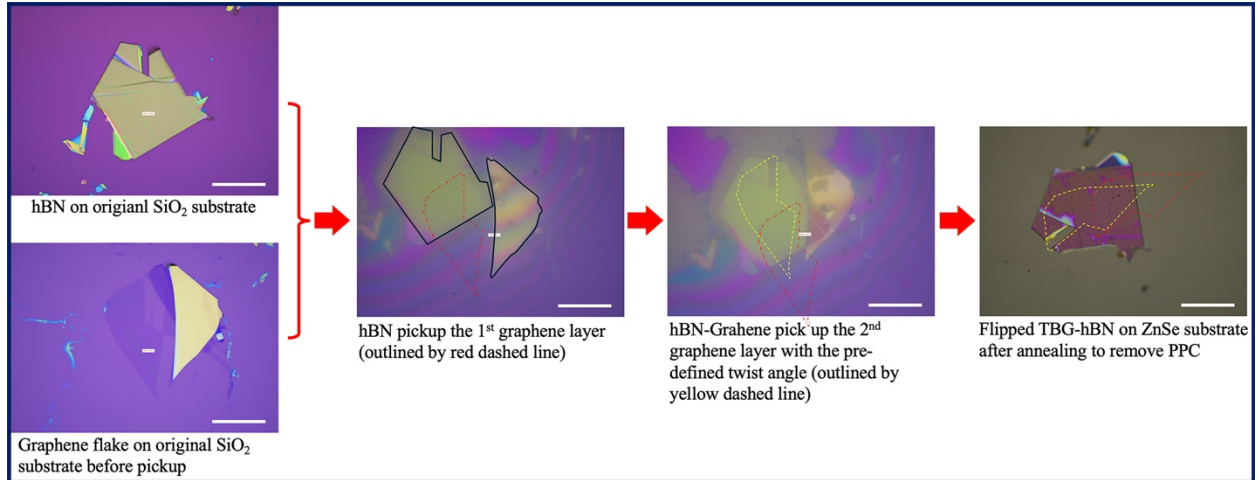
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Supplementary Note 1: Hexagonal Boron Nitride Phonon-Polariton Fitting

The complex wavenumber $\tilde{q}(k)$ of hyperbolic PhPs in hBN is described by the equation:

$$\tilde{q}(\lambda) = -\frac{\sqrt{\epsilon_{\parallel}(\lambda)}}{id\sqrt{\epsilon_{\perp}(\lambda)}} \left[\arctan\left(\frac{i\epsilon_a(\lambda)}{\sqrt{\epsilon_{\parallel}(\lambda)\epsilon_{\perp}(\lambda)}}\right) + \arctan\left(\frac{i\epsilon_s(\lambda)}{\sqrt{\epsilon_{\parallel}(\lambda)\epsilon_{\perp}(\lambda)}}\right) \right] \quad (1)$$

Where $\epsilon_{\parallel}(\lambda)$ and $\epsilon_{\perp}(\lambda)$ are hBN's in and out-of-plane wavelength-dependent permittivity, $\epsilon_s(\lambda)$ is the sample permittivity, $\epsilon_a(\lambda)$ is the air (or top layer's) permittivity, and d is the flake thickness¹. When IR light is coupled into the tip-sample system (hBN flake), propagating PhPs can, in principle, be launched by both the flake edge and the tip, and subsequently detected by the tip²⁻⁴. Compared to edge-launched PhPs with wavelength λ_p , tip-launched PhPs travel twice the distance before detection, resulting in a standing wave with wavelength $\lambda_p/2$. Nevertheless, tip-launched PhPs typically exhibit stronger amplitudes from destructive interference, but rapid optical decay due to the travel distance, making them difficult to observe. Consequently, most PhP measurements in literatures display only a single oscillatory pattern, and detecting both requires either isotopically pure hBN or a more sensitive technique capable of capturing the fast-decaying tip-launched contribution. These two processes can be modeled by exponential decaying sinusoids as expressed in Eqs. 2 and 3:

$$f_{edge-launched}(x) = d_0 + A_1 \cdot \sin\left[\frac{(x-\mu)2\pi}{\lambda_1} + \phi_1\right] \cdot e^{\frac{-(x-\mu)}{\tau_1}} \quad (2)$$

$$f_{edge+tip\,launched}(x) = d_0 + A_1 \cdot \sin\left[\frac{(x-\mu)2\pi}{\lambda_1} + \phi_1\right] \cdot e^{\frac{-(x-\mu)}{\tau_1}} + A_2 \cdot \sin\left[\frac{(x-\mu)2\pi}{\lambda_2} + \phi_2\right] \cdot e^{\frac{-(x-\mu)}{\tau_2}} \quad (3)$$

Where d_0 is the background signal, A is the oscillation amplitude, ϕ is the phase offset, and μ is a constant offset set to the position of the first peak's maximum to ensure consistent phase between fits.

The tapping AFM-IR profile (Supplementary Fig. 1) exhibits a single oscillatory pattern, whereas the TFM-IR profile (Fig. 1c) from hBN flake reveals a distinct beating pattern at the same wavenumber. Fitting these profiles with Eqs. 2 and 3, respectively, yields $\lambda_1 = 373.5 \pm 1.6 \text{ nm}$ for tapping AFM-IR, and $\lambda_1 = 361.9 \pm 1.6 \text{ nm}$ together with $\lambda_2 = 188.3 \pm 1.0 \text{ nm}$ for TFM-IR.

In TFM-IR, the relation $\lambda_1 \approx 2\lambda_2$ is consistent with λ_1 obtained from tapping AFM-IR, confirming their respective physical origins of the two oscillatory components.

Supplementary Note 2: TFM-IR Theory with Non-Linear Frequency Mixing

Supplementary Fig. 3 shows schematics of an AFM cantilever in torsional motion. Torsion is measured according to the cantilever's angle with respect to the rest plane $\Theta(x, t)$, at the position x along the cantilever, and at time t . The dynamics of torsional motion of a bar-like structure has been well studied^{5,6}, and the motion for a cantilever's rotation around its central axis is governed by the equation below:

$$\frac{\partial}{\partial x} \left[G J(x) \frac{\partial \Theta(x, t)}{\partial x} \right] + m_T(x, t) = \rho(x) I(x) \frac{\partial^2 \Theta(x, t)}{\partial t^2} \quad (4)$$

where G is material's shear modulus, and $J(x)$ is the second polar moment of the area, determined according to the equation $\theta = TL/GJ$ relating angle of rotation θ to torque T at the end of a bar of length L . The product $GJ(x) = c_T(x)$ is referred to as torsional stiffness. $m_T(x, t)$ is the external twisting moment per-unit-length acting on the cantilever, $\rho(x)$ is the mass density, and $I(x)$ is the cross-section polar moment of inertia.

For a free-standing AFM cantilever, a constant cross-sectional area $J(x) = J$, and a constant mass density $\rho(x) = \rho$ can be assumed. Eq. 4 is simplified to

$$c_T \frac{\partial^2 \Theta(x, t)}{\partial x^2} - \rho I \frac{\partial^2 \Theta(x, t)}{\partial t^2} = 0. \quad (5)$$

By separating variable $\Theta(x, t) = g(x)\theta(t)$ and defining the eigenvalues as $\eta^2 = \omega^2 \rho I / c_T$, we get:

$$\begin{cases} \frac{\partial^2 g(x)}{\partial x^2} + \eta^2 g(x) = 0 \\ \frac{\partial^2 \theta(t)}{\partial t^2} + \omega^2 \theta(t) = 0 \end{cases} \quad (6)$$

This has led to two simple harmonic oscillators, whose constants A_1, A_2 are set by the cantilever's physical boundary conditions, and B_1, B_2 by the initial conditions with solutions below:

$$\begin{cases} g(x) = A_1 \sin(\eta x) + A_2 \cos(\eta x) \\ \theta(t) = B_1 \sin(\omega t) + B_2 \cos(\omega t) \end{cases} \quad (7)$$

For an AFM cantilever, its base is clamped (Fig. 1a) and the other end is free without any twisting moment. As a result, the boundary condition can be set as:

$$\begin{cases} \Theta(0, t) = 0 \\ c_T \frac{\partial \Theta(x, t)}{\partial x} \big|_{x=L} = 0 \end{cases} \quad (8)$$

Applying them gives the eigenvalue equation $\cos(\eta L) = 0$, which is satisfied for $\eta_n = \left(n - \frac{1}{2}\right) \frac{\pi}{L}$. Substituting this back into Eq. 7, the free spatial eigenmodes are:

$$g_n(x) = \sin \eta_n x ; \quad \int_0^L g_n(x) g_m(x) dx = L \delta_{nm} \quad (9)$$

Correspondingly, the resonant frequency of each eigenmode is:

$$\omega_n = \eta_n \sqrt{\frac{\rho I}{c_T}} \quad (10)$$

This has led to one important difference between a cantilever's torsional and flexural modes, the torsion modes have a linear dispersion while the flexural modes possess a quadratic dispersion as $\omega \propto \eta^2$ (refs. 5-7). This linear dispersion suggests the possibility of non-linear harmonic interactions between different torsional modes.

When the AFM tip is in contact with sample, the tip-sample interaction imposes a twisting moment and yields new boundary conditions. Assuming that the cantilever angle is small, the magnitude of the twisting moment induced by a normal force can be expressed as:

$$m_T = |\vec{r} \times \vec{F}| = HF \sin \theta \approx HF\theta \quad (11)$$

where H is the height of tip (Supplementary Fig. 3). The updated boundary conditions are:

$$\begin{cases} \Theta(0, t) = 0. \\ c_T \frac{\partial \Theta(x, t)}{\partial x} \big|_{x=L} = HF\Theta(L, t) \end{cases} \quad (12)$$

The first condition requires $A_2 = 0$ in Eq. 7. The second boundary condition results in the equation:

$$\begin{cases} c_T A_1 \eta \cos(\eta L) = H F A_1 \sin(\eta L) \\ \tan(\eta L) = \frac{c_T L}{H F} \frac{1}{\eta L} \end{cases} \quad (13)$$

The roots of this transcendental equation are the new η_n . To better understand their behavior, we can combine the tip-sample interaction into a single parameter $q = \frac{FH}{c_T L}$. Considering the limiting cases for q ,

$$\tan(\eta L) = \frac{1}{q} \frac{1}{\eta L} \rightarrow \begin{cases} \eta_n = \left(n - \frac{1}{2}\right) \frac{\pi}{L}, & q \rightarrow 0 \\ \eta_n = n \frac{\pi}{L} & q \rightarrow \infty \end{cases} \quad (14)$$

The roots shift by up-to $\pi/2L$ with increasing contact. These two limiting cases are analogous to standing waves in pipes, where the standing wave ends at an anti-node when the end is open, or a node when the end is closed. This highlights the importance of AFM feedback setting in TFM-IR measurement. The orthogonality property in Eq. 8 is broken with these new η_n , but for the following discussions we will assume that the tip-sample contact is weak enough that the modes are still approximately orthogonal.

In general, the torsional motion of AFM cantilever with tip-sample interaction can be described as:

$$\rho I \frac{\partial^2 \Theta(x,t)}{\partial t^2} - \gamma' \frac{\partial \Theta(x,t)}{\partial t} - c_T \frac{\partial^2 \Theta(x,t)}{\partial x^2} = F(x,t) \quad (15)$$

Where, γ' is the damping coefficient, $F(x,t)$ is the force exerted to the tip and can be further expressed as $F_0 \delta(x-L) f(t)$. The solution Θ can be expanded by using eigenmodes from the Eq.9 as $\Theta(x,t) = \sum_n P_n g_n(x) \theta_n(t)$. By substituting this into Eq.15, we obtain:

$$\sum_n P_n \left[\rho I g_n \frac{\partial^2 \theta_n}{\partial t^2} - \gamma' g_n \frac{\partial \theta_n}{\partial t} - c_T \frac{\partial^2 g_n}{\partial x^2} \theta_n \right] = F_0 f(t) \delta(x-L) \quad (16)$$

By reorganizing the equation and letting $\gamma = \frac{\gamma'}{\rho I}$, the Eq.16 can be further simplified as:

$$\sum_n P_n g_n \left[\frac{\partial^2 \theta_n}{\partial t^2} - \gamma \frac{\partial \theta_n}{\partial t} + \omega_n^2 \theta_n \right] = \frac{F_0}{\rho l} f(t) \delta(x - L) \quad (17)$$

We project this relation onto the m -th mode by integrating both sides of Eq.17, we obtain:

$$\int_0^L dx g_m(x) \sum_n P_n g_n \left[\frac{\partial^2 \theta_n}{\partial t^2} - \gamma \frac{\partial \theta_n}{\partial t} + \omega_n^2 \theta_n \right] = \frac{F_0}{\rho l} f(t) \int_0^L dx g_m(x) \delta(x - L) \quad (18)$$

This can be further simplified as:

$$L P_n \left[\frac{\partial^2 \theta_n}{\partial t^2} - \gamma \frac{\partial \theta_n}{\partial t} + \omega_n^2 \theta_n \right] = \frac{F_0}{\rho l} g_n(L) f(t) \quad (19)$$

By separating the spatial and temporal parts, we get:

$$\begin{cases} P_n = \frac{F_0}{\rho l L} g_n(L) \\ \ddot{\theta}_n - \gamma \dot{\theta}_n + \omega_n^2 \theta_n = f(t) \end{cases} \quad (20)$$

One important implication from Eq. 20 is that under external force excitation, the n -th torsional mode of AFM cantilever can be described as a damped-driven harmonic oscillator. This provides the basis for configuring different measurement schemes in TFM-IR.

We have evaluated one experimental TFM-IR scheme outlined in Fig. 1, where an AFM cantilever is driven at its inherent m -th torsional mode while monitoring the dynamics of its orthogonal non-driven n -th mode under optical excitation by a pulse laser at a repetition rate f_{laser} . This results in an effective force $f(t) = F_{eff}(t)$ in Eq. 20 exerted to the tip, which provides momentum for torsional motion of cantilever, involving photothermal induced force.

When the cantilever is strongly driven at the m -th torsional mode with large angular amplitude A_m , the total tip angle in Fig. 1b can be written as:

$$\theta(t) = \theta_n(t) + A_m \cos \omega_m t \quad (21)$$

where θ_n is the contribution from the inherent non-driven n -th mode of the cantilever,

with $\theta_n \ll A_m \cos \omega_m t$. As shown in Fig.1b, the optical induced force $\vec{F}(f_{laser}, t)$ can be decomposed to its in-plane ($\vec{F}_{in}$) and out-of-plane ($\vec{F}_{out}$) components determined by the anisotropic characteristics of sample, and only their partial components normal to the tip can contribute to the momentum for torsional motion of cantilever. $F_{eff}(t)$ can thus be expressed:

$$\begin{aligned} F_{eff}(t) &= F_{in}(f_{laser}, t) \cos[\theta_n + A_m \cos(\omega_m t)] + F_{out}(f_{laser}, t) \sin[\theta_n + A_m \cos(\omega_m t)] \\ &\approx F_{in}(f_{laser}, t) \cos[A_m \cos(\omega_m t)] + F_{out}(f_{laser}, t) \sin[A_m \cos(\omega_m t)] \end{aligned} \quad (22)$$

For simplicity, we can write the periodic laser-induced force as $F_{in}(f_{laser}, t) = F_{in} \cos(2\pi f_{Laser} t)$ and $F_{out}(f_{laser}, t) = F_{out} \cos(2\pi f_{Laser} t)$. By employing the Jacobi-Anger expansion, we can have:

$$\begin{cases} \sin(A_m \cos \omega_m t) = 2 \sum_{k=0}^{\infty} (-1)^k J_{2k+1}(A_m) \cos((2k+1)\omega_m t) \\ \cos(A_m \cos \omega_m t) = J_0(A_m) + 2 \sum_{k=1}^{\infty} (-1)^k J_{2k}(A_m) \cos((2k)\omega_m t) \end{cases} \quad (23)$$

where $J_k(A_m)$ is the Bessel function of order k .

Then the effective force becomes:

$$\begin{aligned} F_{eff}(t) &= F_{in} \cos(2\pi f_{Laser} t) (J_0(A_m) + 2 \sum_{k=1}^{\infty} (-1)^k J_{2k}(A_m) \cos((2k)\omega_m t)) + \\ &\quad F_{out} \cos(2\pi f_{Laser} t) (2 \sum_{k=0}^{\infty} (-1)^k J_{2k+1}(A_m) \cos((2k+1)\omega_m t)) \end{aligned} \quad (24)$$

By using trigonometric product identity and letting $\omega_m = 2\pi f_m$, we have:

$$\begin{aligned} F_{eff}(t) &= F_{in} J_0(A_m) \cos(2\pi f_{Laser} t) + F_{in} \sum_{k=1}^{\infty} (-1)^k J_{2k}(A_m) (\cos(2\pi(f_{Laser} + 2kf_m)t) + \\ &\quad \cos(2\pi(f_{Laser} - 2kf_m)t)) + F_{out} \sum_{k=0}^{\infty} (-1)^k J_{2k+1}(A_m) (\cos(2\pi(f_{Laser} + (2k+1)f_m)t) + \\ &\quad \cos(2\pi(f_{Laser} - (2k+1)f_m)t)) \end{aligned} \quad (25)$$

Projecting expression of $F_{eff}(t)$ in Eq.25 onto Eq.20, we have immediately achieved a few important findings:

(1) By driving the cantilever's m -th torsional mode at large amplitude A_m , nonlinear mixing can occur between the laser-induced force and the torsional motion, and the

non-driven n -th torsional mode ($\omega_n = 2\pi f_n$) can be resonantly excited if the frequency mixing between f_{laser} and f_m coincide with f_n within the linewidth: $f_n = f_{laser} \pm 2kf_m$, or $f_n = f_{laser} \pm (2k - 1)f_m$, ($k = 1, 2, 3, \dots$).

(2) Importantly, the in-plane (F_{in}) and out-of-plane (F_{out}) force components can be selectively and resonantly coupled to the non-driven n -th torsional mode, by judiciously tuning the f_{laser} to be $f_n \pm 2kf_m$ (even-harmonic mixing) or $f_n \pm (2k - 1)f_m$ (odd-harmonic mixing), respectively. This can therefore provide a simple but flexible method to separately measure anisotropic in-plane and out-of-plane sample properties while maximizing signal-to-noise ratio through resonant amplification of the targeted torsional mode, which has been challenging otherwise. In essence, the combination of an m -th mode torsional drive with large-amplitude, orthogonal eigenmodes, and nonlinear force projection in TFM-IR enables a tunable frequency mixer that converts optical responses into a resonantly detectable n -th mode torsional response.

(3) The amplitude of different frequency mixing is weighted by the Bessel function, $J_k(A_m)$ which decrease with order k . Interestingly, we have found that for the detection of the first order of in-plane ($f_n = f_{laser} \pm 2f_m$) and out-of-plane ($f_n = f_{laser} \pm f_m$) force components, their corresponding signal amplitudes scale differently as:

$$\begin{cases} f_n = f_{laser} \pm f_m & \Rightarrow & \frac{F_{out}}{2} A_m \propto A_m \\ f_n = f_{laser} \pm 2f_m & \Rightarrow & \frac{F_{in}}{8} A_m^2 \propto A_m^2 \end{cases} \quad (26)$$

This has been confirmed by our experiment (Supplementary Fig. 5).

(4) In principle, if laser repetition rate is set to have $f_{laser} = f_n$, the direct term involving F_{in} in Eq. 25 is always present, however this signal is very small and not augmented by A_m , and the background noise or mechanical noise/electrical noise can reduce signal-to-noise ratio substantially, making the detection with this scheme challenging in experiment.

Supplementary Note 3: Strain-Modified Photothermal Effect

The TFM-IR images of mica nanobubbles in Fig. 2a show distinct annular features that differ markedly from their height profiles. In general, the absorption of IR light by the sample induces a local temperature increase, producing a photothermal effect. The resulting mechanical expansion exerts a force on the AFM tip, giving rise to the TFM-IR signal^{7,8}. The disparity between the TFM-IR and morphological maps immediately suggests inhomogeneous thermo-mechanical response within the nanobubble. The TFM-IR observable can be effectively expressed as:

$$S(\vec{r}, \lambda, \omega_n) \propto \sigma_{IR}(\vec{r}, \lambda) \Delta L(\vec{r}) \chi(\omega_n) \quad (27)$$

where σ_{IR} is the local optical absorption, ΔL is photothermal expansion caused by local thermal rise, and χ is the mechanical transfer function determined by the torsional dynamics of the cantilever at the n -th detection mode, while accounting for tip-sample interactions as detailed in Supplementary Note 2. The local photothermal expansion $\Delta L(\vec{r})$ is given by⁹:

$$\Delta L(\vec{r}) = \alpha(\vec{r}) L_0(\vec{r}) \Delta T(\vec{r}) \quad (28)$$

where α is the local thermal expansion coefficient, L_0 is the effective length along measurement direction, and ΔT is the local temperature rise due to optical absorption. Suppose the sample has a local strain $\epsilon(\vec{r})$ such that $L'_0 = L_0(1 + \epsilon)$, then under the same temperature change ΔT , the strain modified expansion becomes $\Delta L' = L'_0 \alpha \Delta T$, leading to:

$$\Delta L'(\vec{r}) = (1 + \epsilon(\vec{r})) \alpha(\vec{r}) L_0(\vec{r}) \Delta T(\vec{r}) \quad (29)$$

Therefore, a locally stretched region appears to have a slightly larger effective thermal expansion, while a compressed region appears smaller. The effective thermal expansion coefficient $\alpha' = (1 + \epsilon) \alpha$ describes how information about the sample's strain-induced displacement is embedded within the TFM-IR's $S(\vec{r}, \lambda, \omega_n)$ signal by effectively modifying the thermal expansion coefficient. It is important to note that if the strain is anisotropic (as in the case of mica nanobubble), directional coefficients α_i need to be considered alongside the local strain tensor ϵ_{ij} . This leads naturally to differences between in-plane and out-of-plane TFM-IR mapping. In addition to this dominant strain-induced modification of the thermal expansion, there are a few other possible effects

under large strain, including strain-induced modification of optical absorption $\sigma_{IR}(\vec{r}, \lambda)$ via the photoelastic effect, strain-induced shifts in the band structure, phonon frequencies, or refractive index.

Supplementary Note 4: TFM-IR Image Reconstructions for Mapping Anisotropic Strain Distribution

For the mica nanobubble in Fig. 2a, the dome-shaped geometry naturally generates inhomogeneous strain, modifying local thermal coefficient and amplifying photothermal expansion under pulsed IR excitation in these regimes. Therefore, qualitatively the TFM-IR images provide a direct and distinctive fingerprint of the nanobubble's inhomogeneous strain distribution. In particular, the nonlinear frequency mixing schemes unique to TFM-IR, as described in Supplementary Note 2, enable direct mappings of both in-plane and out-of-plane strain components.

By performing TFM-IR spectra with different frequency mixing scheme, we have confirmed the capability to detect in-plane and out-of-plane vibrational modes, as presented in Fig. 2a. However, cross-talking between the in-plane and out-of-plane response is also observed. For example, the out-of-plane vibrational signal does not vanish in the in-plane detection, and vice versa. Such crosstalk between the two channels arises from several physical and instrumental factors. For example, as discussed in Supplementary Note 2, the assumption of complete decoupling between torsional modes is not strictly valid in practice: tip-sample contact inevitably induces coupling not only between torsional modes but also between torsional and flexural modes. Geometric asymmetries of the tip or imperfect tip mounting in holder can further exacerbate this coupling. in the tip holder. Furthermore, while our model treats the AFM tip as a point detector, in reality it has a finite contact area. Thus, when tip contacts sample, the detected signal represents a weighted average of the photothermal expansion across the contact area. A vertical expansion on one side of the tip can induce a torque, producing torsional deflection that partially projects onto the in-plane measurement channel. Similarly, a lateral expansion can produce a small displacement component contributing to the out-of-plane channel. Additionally, imperfections in quadrant photodetector alignment or the finite size of laser spot on the PSD (Fig. 1a) can further mix the measured signals. Together, these factors account for the leakage between the two demodulated channels.

Collectively, these effects can be represented by a linear instrument response matrix $\mathbf{M}$, which quantifies the leakage between in-plane and out-of-plane measurements. We define the matrix $\mathbf{M}$ as the linear transformation between the measured TFM-IR signal ($\mathbf{I}$) and real signal ($\mathbf{S}$):

$$\begin{bmatrix} I_{in}(\lambda) \\ I_{out}(\lambda) \end{bmatrix} = \begin{bmatrix} M_{in}^{in} & M_{in}^{out} \\ M_{out}^{in} & M_{out}^{out} \end{bmatrix} \begin{bmatrix} S_{in}(\lambda) \\ S_{out}(\lambda) \end{bmatrix} \quad (30)$$

where $I_{in}(\lambda)$ and $S_{in}(\lambda)$ are measured and real signal at wavelength λ in an in-plane measurement, and $I_{out}(\lambda)$ and $S_{out}(\lambda)$ are measured and real signal at wavelength λ in an out-of-plane measurement. The diagonal entries of $\mathbf{M}$ (M_{in}^{in} , M_{out}^{out}) encode channel gains (i.e., how much of each true signal gets detected to its corresponding measurement), and the off-diagonal entries of $\mathbf{M}$ (M_{in}^{out} , M_{out}^{in}) encode cross-talk components. By choosing and comparing intensities of two characteristic in-plane and out-of-plane markers, λ_{in} and λ_{out} , respectively, in each measurement, matrix $\mathbf{M}$ can be evaluated. Once $\mathbf{M}$ is calibrated, related crosstalk can be corrected to reconstruct anisotropic photothermal maps with more accurate information.

If the characteristic λ_{in} and λ_{out} are well separated in energy, we can assume $S_{in}(\lambda_{out}) \ll S_{out}(\lambda_{out})$ and $S_{out}(\lambda_{in}) \ll S_{in}(\lambda_{in})$. The matrix $\mathbf{M}$ can thus be determined by performing in-plane and out-of-plane spectra covering the wavelength λ_1 and λ_2 . We further define the crosstalk ratio R as:

$$\begin{cases} R_{out} = \frac{M_{in}^{out}}{M_{out}^{out}} \\ R_{in} = \frac{M_{out}^{in}}{M_{in}^{in}} \end{cases} \quad (31)$$

This provides a pathway to reconstruct genuine in-plane RC_{in} and out-of-plane RC_{out} maps from the measured (or “raw”) in-plane (C_{in}) and out-of-plane (C_{out}) maps by scaling the measured image pixel-by-pixel to match the amplitude of the cross-talk signal as determined by Eq.31. This reconstruction is expressed by using inverting matrix $\mathbf{M}^{-1}$ as:

$$\begin{bmatrix} RC_{in} \\ RC_{out} \end{bmatrix} = \mathbf{M}^{-1} \begin{bmatrix} C_{in} \\ C_{out} \end{bmatrix} = \frac{1}{M_{in}^{in}M_{out}^{out} - M_{in}^{out}M_{out}^{in}} \begin{bmatrix} M_{out}^{out} & -M_{in}^{out} \\ -M_{out}^{in} & M_{in}^{in} \end{bmatrix} \begin{bmatrix} C_{in} \\ C_{out} \end{bmatrix} \quad (32)$$

Therefore, the reconstructed RC_{in} and RC_{out} can be obtained:

$$\begin{cases} RC_{in} = \frac{C_{in} - R_{out}C_{in}}{M_{in}^{in}(1 - R_{in}R_{out})} \\ RC_{out} = \frac{C_{out} - R_{in}C_{in}}{M_{out}^{out}(1 - R_{in}R_{out})} \end{cases} \quad (33)$$

For the mica nanobubble shown in Fig. 2, we selected $\sim 995 \text{ cm}^{-1}$ and 1050 cm^{-1} as the out-of-plane and in-plane markers, respectively, due to their well-separated energies. By comparing their relative intensities in the f_{out} - and f_{in} - spectra, we obtained $R_{in} = 0.814$ and $R_{out} = 0.538$. These values form the basis for image reconstruction aimed at minimizing crosstalk, and the results are presented in Figs. 2b and 3.

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