Dispersive heterodyne Brillouin spectroscopy

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Frequency- and time-domain Brillouin spectroscopy\textsuperscript{1,2} are powerful tools to read out the mechanical properties of complex systems in material\textsuperscript{3,4} and life sciences\textsuperscript{5,6}. Indeed, coherent acoustic phonons in the time-domain method offer superior depth resolution and a stronger signal than incoherent acoustic phonons in the frequency-domain method. However, it does not allow multichannel detection and therefore falls short in signal acquisition speed. Here, we present Brillouin spectroscopy that spans time- and frequency-domain to allow multichannel detection of Brillouin scattering light from coherent acoustic phonons. Our technique maps time-evolve Brillouin oscillations in a chromatic-dispersed laser pulse’s instantaneous frequency. Spectroscopic heterodyning of the Brillouin oscillations in the frequency domain enhances signal acquisition speed by at least 100-fold over the time-domain method. As a proof of concept, we imaged heterogeneous thin films over a wide bandwidth with nanometer depth resolution. We, therefore, foresee that our approach catalyses future phonon spectroscopy toward real-time mechanical imaging.

Characterization of the micro/nanoscale material’s mechanical properties is critical for understanding their functionality\textsuperscript{7,8}. Researchers have been looking for a means to measure on a tiny scale for several decades, but there are only a few tools for obtaining the mechanical properties of miniaturised samples. Common mechanical tests (e.g., dynamic mechanical analysis and rheometer) for bulk materials require contact mechanical forces to samples and are difficult to scale down their size to obtain small-scale spatial information. Although atomic force microscopy\textsuperscript{9,10} enables two-dimensional measurements with a nanometre resolution, it is limited to a surface measurement. Thus, despite the increasing advancement of miniaturised mechanical testing, it is an open challenge to realize in-depth non-contact microscopic measurements.
An acousto-optical technique can meet this demand; Brillouin spectroscopy interrogates material’s mechanical properties from an inelastic light scattering by a phonon, Brillouin scattering\textsuperscript{11}. It non-invasively reads out the viscoelastic properties of small-scale materials with high spatial resolution. Brillouin spectroscopy has various applications, including phonon physics studies\textsuperscript{12,13} and characterisation of solids\textsuperscript{14,15}, liquids\textsuperscript{16,17} and biological samples\textsuperscript{18}. In general, Brillouin spectroscopy can be classified into two forms: (1) as frequency-domain Brillouin spectroscopy, which records GHz frequency-shifted Brillouin scattering light from incoherent acoustic phonons by a specific imaging spectrometer (e.g., virtually imaged phased array spectrometer\textsuperscript{19}), and (2) as time-domain Brillouin spectroscopy, which records carrier frequency of Brillouin scattering light from coherent acoustic phonons, known as a Brillouin oscillation, by picosecond ultrasonics\textsuperscript{20,21} based on a step-scan pump-probe method with a single photodetector.

In this Letter, we propose and demonstrate Brillouin spectroscopy that straddles the time and frequency domain, which we refer to as dispersive heterodyne Brillouin spectroscopy. Our technique harnesses coherent acoustic phonons and a chirped laser pulse to record time-evolving Brillouin oscillations in the frequency domain. Coherent acoustic phonons launched from the metallic thin film provide nanometre depth resolution and intense Brillouin scattering light as in the time-domain Brillouin spectroscopy. The chirped laser pulse with a linear chromatic dispersion gives the single shot and multichannel detection of Brillouin scattering light as in the frequency-domain Brillouin spectroscopy. Accordingly, our technique inherently possesses higher depth resolution and better signal-to-noise ratio than conventional frequency-domain Brillouin spectroscopy and at least 2-orders faster signal acquisition speed than conventional time-domain Brillouin spectroscopy. Our dispersive heterodyne Brillouin spectroscopy is as versatile as
conventional Brillouin spectroscopy holding great promise as a practical microscopic mechanical imaging modality.

In our technique of dispersive heterodyne Brillouin spectroscopy, we use a single ultrafast short pulse laser to pump and probe coherent acoustic phonons (Fig. 1a). The coherent acoustic phonons are launched into the sample by the absorption of an optical pump pulse and subsequent rapid thermal expansion of a metallic thin film, which acts as a transducer (Fig. 1b). A femtosecond laser pulse is temporally stretched by the pulse stretcher (the left inset in Fig. 1a) and incident on the sample as successive probes diffracted by propagating coherent acoustic phonons in different timing and at different positions (Fig. 1b). Heterodyning of the diffracted chirped laser pulse from coherent acoustic phonons and the reflected chirped laser pulse from the transducer surface results in GHz-beating of light intensity in the time domain. A spectrometer (the right inset in Fig. 1a) disperses the chirped laser pulse in the frequency domain to create a Brillouin interferogram (Fig. 1c). The frequency \( f_B \) of these oscillations is related to the acoustic velocity \( v \), the refractive index \( n \), the probe wavelength \( \lambda \) through the relation \( f_B = 2nv/\lambda \) for normal probe beam incidence. The Brillouin interferograms recorded by the image sensor are processed on the computer to construct a time trace of transient reflectivity with the time axis calibrated from the optical settings (Fig. 2a, Supplementary Fig. 1). High sampling frequency (4.2 THz) and ample time window (282 ps) can be achieved with large data points (1024 points). The temporal resolution of the system is determined by the sub-pulse duration \( \tau_c \) given by \( (\tau_0 \tau_c)^{1/2} \) where \( \tau_0 \) is the Fourier transform-limited pulse duration and \( \tau_c \) is the chirped pulse duration. The maximum time window is traded with a temporal resolution, where increasing the chirp elongates the time window but degrading temporal resolution (Supplementary Fig. 2). Our setup used a chirped pulse of full-width half-maximum (FWHM) temporal duration of 160 ps and 400 nm-centred wavelengths with an FWHM bandwidth
of 7.8 nm corresponding to 31 fs transform-limited pulse duration. From these parameters, our current system has a temporal measurement resolution of 2 ps.

To gauge the performance of dispersive heterodyne Brillouin spectroscopy, we measured the Brillouin frequency of a silicon dioxide (SiO$_2$) thin film attached to a 100 nm chromium film (Fig. 2b). We used SiO$_2$ thin film as a sample for our proof-of-principle demonstration because the acoustic velocity ($v \approx 5.9$ km/s), refractive index ($n \approx 1.5$), and Brillouin frequency ($f_B \approx 44$ GHz) at $\lambda \approx 400$ nm are reported in previous studies$^{23,24}$. The time trace of Brillouin oscillations (Fig. 2c, Supplementary Fig. 3) and the Brillouin spectrum (Fig. 2d) obtained using our technique (4000 shots averaging) show good agreement with those measured by time-domain Brillouin spectroscopy (step-scan method). The calculated thickness of SiO$_2$ thin film from a bump on time trace around 180 ps originating from SiO$_2$–air interface is 1 µm which matches the actual thickness of SiO$_2$. Moreover, we showed that our technique can measure Brillouin frequency of liquid and gel samples (Supplementary Fig. 4). Meanwhile, there were slight differences in Brillouin oscillation amplitude and a non-oscillatory signal component due to thermally induced changes in reflectivity of chromium film originating from indispensable wavelength dependence on piezo-optical couplings$^{25}$. It is also worth mentioning that there is much interest in the Brillouin frequency than the transient changes of the refractive index in the transducer which are routinely removed$^{26}$.

We show a nanometre-resolution depth profiling of the heterogeneous thin film consisting of silicon nitride (Si$_3$N$_4$) and SiO$_2$ (Fig. 3a); Si$_3$N$_4$ is widely used in opto-electrical devices, and it has an approximately three times higher Young modulus than SiO$_2$$^{24}$. From the time trace in Fig. 3b, one can recognise the change in a phase-matching condition of probe light and coherent acoustic phonons when the coherent acoustic phonons leave Si$_3$N$_4$ entering SiO$_2$ (Fig. 3b). The
high-frequency oscillations \( (f_B = 90 \text{ GHz}) \) from the Si\(_3\)N\(_4\) layer firstly appear, and the low-frequency oscillations \( (f_B = 44 \text{ GHz}) \) start with a delay of 80 ps, which corresponds to the crossing of coherent acoustic phonons at the interface of the SiO\(_2\) layer (Fig. 3c). The Si\(_3\)N\(_4\) has a higher Brillouin frequency than SiO\(_2\), as expected, because the product of the material’s refractive index and acoustic velocity differ by a factor of two. We also performed depth profiling of 1µm SiO\(_2\) step created on titanium film (Fig. 3d). We obtained spatial profile by translating the sample over a 3 mm with 100 µm pitch. From the transient reflectivity map shown in Fig. 3e, we can observe Brillouin oscillations from the SiO\(_2\) layer until a lateral position at 2.2 mm. The thickness of SiO\(_2\) thin film (Supplementary Fig. 5) corresponds well to the bumps on the time trace around 150 ps originating from the SiO\(_2\)–air interface. The lateral boundary of the film becomes apparent near 1.2 mm, and the thickness of the film decreases as it approaches the bare titanium region (Fig. 3f).

To demonstrate a Brillouin imaging capability, we used glycerol, a well-known and well-characterised prototypical glass-forming liquid\(^ {27} \), squeezed between two flat glass substrates; one of them is a titanium film with 1 µm SiO\(_2\) step. To obtain the transient reflectivity map, the sample was translated with a 200 µm-step over 6 mm (Fig. 4a). A total of 31 waveforms were acquired, with 4000 shots averaging per step. We can observe the interface between SiO\(_2\) and glycerol from the contrast produced by the Brillouin frequency (Fig. 4b). A spatial variation in the Brillouin frequency is mapped in Fig. 4c.

This work has demonstrated the feasibility of dispersive heterodyne Brillouin spectroscopy, which combines two key mechanisms—localised spectral encoding of the Brillouin oscillation and decoding by spectrometer—to perform single-shot multichannel detection of Brillouin oscillations. As a result, our technique can acquire Brillouin oscillations at least 100-times faster than the step-scan method in time-domain Brillouin spectroscopy. The above
comparisons were performed under the following conditions: the same repetition rate laser, measurement time window, temporal resolution, and an optical delay line with an infinitely short positioning time. While the frame interval of the image sensor used as a multichannel detector limits the overall measurement time of the current proof-of-principle experiment, this limitation can be addressed through further system improvements, such as using a high-frame-rate camera with a higher repetition rate laser source. Further improving acquisition rate, e.g., by changing the operating wavelength to the near-infrared region to employ dispersive Fourier-transform spectroscopic techniques\textsuperscript{28}, can profoundly benefit translating our technique toward a study on moving biological specimens and flow cytometric applications. Thus, its potential applications are versatile and introduce a new paradigm in Brillouin spectroscopy to open a door for the prospect of the unexploited fields of material and life sciences.

**Methods**

**Brillouin spectroscopy system**

The ultrashort laser source we used is a Ti:sapphire regenerative amplifier (Astrella-USP-1K, Coherent, US) producing 803 nm-centered wavelengths with a bandwidth of 70 nm, and a pulse duration of 35 fs amplified pulses in a maximum repetition rate of 1 kHz. Because the sensor readout used in the spectrometer led to slow data acquisition, the pulse repetition rate was down counted from 1 kHz to 100 Hz. The laser output was frequency-doubled (406 nm-centred wavelengths with a bandwidth of 7.8 nm) by $\beta$-barium borate crystal (BBO-1001H, Eksma Optics, LT) and separated from 800 nm laser pulse by a harmonic separator (USB21, Thorlabs, US) to use as a probe pulse.
An 800 nm fs-laser pulse, modulated at 50 Hz by an optical chopper (MC2000B, Thorlabs), served as a pump pulse to excite coherent acoustic phonons. A probe pulse is time-stretched to 160 ps in FWHM by a pair of 36000 lines/mm gratings (PC3600, Spectrogon, SE). Afterward, the probe pulse is split into a sample and a reference arm by a beam splitter (BS013, Thorlabs). After passing via a half-wave plate (WPH10M-405, Thorlabs), polarising beam splitter (PBSW-405, Thorlabs), and a quarter-wave plate in the sample arm, a lens \( f = 40 \) mm focuses a probe pulse at the sample (SAQWP05M-700, Thorlabs). The probe pulse reflected from the sample, and a fraction of probe pulse that bypassed the sample was directed into a homebuilt Czerny–Turner spectrometer (spectral resolution of 0.037 nm, pixel resolution of 0.0169 nm/pixel, spectral bandwidth of 17.3 nm) coupled to a 16-bit sCMOS camera (Orca Flash4.0 V3, Hamamatsu Photonics, JP) for realising balanced detection. Our spectrometer consists of two spherical mirrors (CM508-500-F01, Thorlabs) and 2400 lines/mm grating (GH50-24V, Thorlabs). Using a digital delay generator (DG645, Stanford Research Systems, US), the camera and chopper were synchronised to the laser’s repetition rate.

**Transient reflectivity calculation**

Transient reflectivity waveforms were constructed from the captured images, yielding a sensitivity of approximately \( 10^{-5} \) with \( 10^3 \) laser shots (Supplementary Fig. 6) by following procedures. First, we excluded pixels at or near the boundary of signal and reference regions. Second, each region corresponding to a specific delay time was averaged vertically over the roughly 500 pixels. Finally, we normalized the value based on signal \( (I_{sig}) \) and reference \( (I_{ref}) \) spectrums recorded with \( (w) \) and without \( (wo) \) the pump pulse:

\[
\frac{\Delta R}{R} = \frac{I_{sig}^{w}}{I_{ref}^{w}} \frac{I_{sig}^{wo}}{I_{ref}^{wo}} - 1
\]
Brillouin imaging

Transient reflectivity waveforms at each measurement position of the sample were acquired by translating a stage attached to the sample mounting holder. We produced a Brillouin frequency map by processing each waveform with a low-pass filter and continuous wavelet transform to obtain a peak Brillouin frequency $f_B$ at different timing. The thickness profile of thin film was constructed by calculating the acoustic velocity $v$ from the Brillouin frequency map through the relation of $v = f_B \lambda / 2n$. The refractive index $n$ of each sub-pulse wavelength $\lambda$ was calculated from the dispersion formula. The timing where coherent acoustic phonons reached the interface between titanium and SiO$_2$ ($T_1$) and SiO$_2$ and its surface ($T_2$), was defined as the jumps in the transient reflectivity waveforms. The spatial profile of the film thickness was calculated by time integration of the acoustic velocity $v$ between $T_1$ and $T_2$ at each measurement position.

Data availability

All relevant data are available from the corresponding author upon reasonable request.

Code availability

The Matlab code used for analysing the data of this study is available from the corresponding author upon reasonable request.
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**Author contributions**

A.I. conceived the concept and designed the systems. A.I. and S.O. constructed the systems and carried out experiments and analyses. I.S. and K.N. provided assistance in constructing the systems, experiments and analysis. A.I. supervised the work and wrote the manuscript. All authors contributed to and approved the final manuscript.

**Competing interests**

The authors declare no competing financial interests.
**Figures**

Figure 1 | Schematic and conceptual illustration of dispersive heterodyne Brillouin spectroscopy. **a**, Schematic of the experimental setup. A near-infrared femtosecond laser pulse is loosely focused on a transducer surface to excite coherent acoustic phonons. A frequency-doubled laser pulse is stretched in the grating pair (left inset) and directed to a spectrometer (right inset) as
a signal after the sample is illuminated. A fraction of the chirped laser pulse that bypassed the sample is also directed into the spectrometer as a reference (light path is not shown in the illustration of the right inset) for balanced detection. b, Principle of dispersive heterodyne Brillouin spectroscopy. Each sub-pulse of the chirped laser pulse (wavelength at \(\lambda_1\) and \(\lambda_2\)) is diffracted by the coherent acoustic phonons at different timings (time at \(T_1\) and \(T_2\)) and in different positions. c, Spectroscopic heterodyning of chirped laser pulse gives the interferogram in a frequency domain that beats at Brillouin frequency.
Figure 2 | Basic performance of dispersive heterodyne Brillouin spectroscopy. a, Time window defined by measuring seven transient reflectivity waveforms of 100 nm chromium thin film with changing the pump delay time by stepping an optical delay line set in the pump arm. b, Schematic of the experiment with the 1 μm SiO₂ film sputtered on a 100 nm chromium film that launched coherent acoustic phonons through transient absorption of an optical pump pulse (1.2 mJ/cm²). The pump pulse excites the coherent acoustic phonons from the glass substrate side, which plays the role of the transducer. A chirped probe pulse then detected acoustic propagation in SiO₂. c, Brillouin oscillations acquired by dispersive heterodyne Brillouin spectroscopy (blue) and time-domain Brillouin spectroscopy with step-scan method (red). d, The Fourier transform of the measured Brillouin oscillations showed a peak at 44 GHz.
Figure 3 | Depth profiling by dispersive heterodyne Brillouin spectroscopy. a, Schematic of the experiment with the 700 nm Si₃N₄ film and 500 nm SiO₂ film sputtered on a 100 nm chromium film. b, Recorded Brillouin oscillations and c, the corresponding time evolution of the Brillouin frequency showing a peak around 90 GHz and 44 GHz. d, Schematic of the sample with 1 μm SiO₂ step created on 300 nm titanium film. e, Recorded Brillouin oscillations and f, the SiO₂ height at each scanning position. Error bars representing the measurement uncertainty (±12 nm error from the temporal resolution and the acoustic velocity of the material) are not visible in the plot.
Figure 4 | Brillouin imaging by dispersive heterodyne Brillouin spectroscopy. a, Recorded Brillouin oscillations and b, corresponding time evolution of the Brillouin frequency at the lateral position of 0 mm. The sample is glycerol, which is squeezed between two flat glass substrates, one of them being coated with a 1 μm SiO₂ film and 300 nm titanium film. c, Brillouin frequency map of the sample showing a signal from SiO₂ (green) and glycerol (blue). The difference in the Brillouin frequencies of the two materials produces a high contrast in the Brillouin image.
Supplementary information

Dispersive heterodyne Brillouin spectroscopy

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Supplementary Figure 1 | Characterisation of the time window. a, Schematic of the experiment with the 100 nm chromium film irradiating 3.6 mJ/cm² optical pump pulse. b, Transient reflectivity waveforms with changing the pump delay time by stepping an optical delay line set in the pump arm.

Supplementary Figure 2 | Dependence of the chirped pulse duration and initial Fourier transform-limited (FTL) pulse duration to the system’s temporal resolution (the sub-pulse duration). The temporal resolution can be tuned to optimise the performance, e.g., by enlarging the bandwidth of the probe pulse.
Supplementary Figure 3 | Recorded transient reflectivity waveform of 1 μm SiO₂ film sputtered on a 100 nm chromium film with different averaging conditions. A series of transient reflectivity recorded from the same region of the sample shows Brillouin oscillation of SiO₂ with an enhancement of signal-to-noise ratio level while increasing the number of shots to be averaged.
**Supplementary Figure 4 | Dispersive heterodyne Brillouin spectroscopy of liquid and gel.**

**a,** Schematic of the experiment with glycerol and hydrogel, which is squeezed between two flat glass substrates, one of them being coated with a titanium thin film. The hydrogel was made of 40% polyacrylamide solution, tetramethyl ethylenediamine, and distilled water at a volume ratio of 25:1:75. The hydrogel solution was fixed at 0.002% (v/v) of ammonium persulfate. **b and d,** Recorded Brillouin oscillations and **c and e,** corresponding spectra for glycerol and hydrogel showing peaks at 20 and 10 GHz, respectively.
Supplementary Figure 5 | Height profile of SiO\textsubscript{2} step measured by a stylus profilometer. The peaks shown in the profile are artifacts originating from the dust on the sample.

Supplementary Figure 6 | Characterisation of the sensitivity. a, The root-mean-square (RMS) fluctuation value is defined by the standard deviation of the transient reflectivity waveform collected without irradiating pump pulses. As the number of averaged shots $N$ increases, the RMS noise level decreases approximately $N^{1/2}$ because laser shot-to-shot fluctuations are random. b, Representative transient reflectivity waveform without pump pulses using 4000-shots of probe pulses for averaging.