Additive Laser Excitation of Multiple Surface Acoustic Waves Up to the Nonlinear Shock Regime

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We introduce a non-destructive method for laser-shock wave generation based on additive superposition of multiple laser-excited strain waves. This technique enables strain generation up to mechanical failure of the sample at pump laser fluences below material ablation or melting thresholds. We demonstrate the ability to generate nonlinear surface acoustic waves (SAWs) in Nb:SnTiO₃ substrates, at typically 1 kHz repetition rate, with associated strains in the percent range. This study paves the way for the investigation of a host of high-strength SAW-induced phenomena, including phase transitions, fatigue, chemistry, and other effects in bulk samples, thin layers, or two-dimensional materials.

Surface acoustic waves involving nanometer-scale atomic motions are used with routine, low-power operation in a plethora of microelectronic devices [1]. The confinement of strain near the sample surface and propagation over millimeter distances make SAWs attractive for a wide range of applications. Typically, these MHz-frequency waves are coherently generated and detected by interdigital microwave transducers (IDTs) deposited on a piezoelectric layer or substrate. Though well-established IDT SAW techniques are limited by small strain amplitudes, it is already possible to employ them to tune material properties [1–5]. Reaching much higher SAW strain amplitudes, up to the mechanical failure of a sample, would open a wide range of novel applications. Until now, pulsed lasers have been used to generate high-amplitude nonlinear SAW shocks with strain amplitudes in the percent range and pressures in the GPa range [6–9], but only operating on a single-shot basis because optical damage to the irradiated sample region which occurs even if the shock itself does not cause damage where it propagates.

A great many phenomena in materials science and solid-state physics and chemistry are mediated by strain, including phase transitions in conventional and quantum materials, plasticity and myriad material failure modes, and surface reconstruction and chemistry. Large static strains of several percent can be applied to bulk or thin samples up to the onset of plasticity or fracture [10–14]. However, since kinetic effects can lead to distinct differences from quasi-static behavior, approaches capable of generating fast actuation mechanisms to explore material responses to dynamic strain are of keen interest. For that purpose, laser-based methods for generation of shock waves have been developed for the study of samples under intense dynamic strain loading. In the most common configuration, bulk shock waves are studied, sometimes non-destructively from femtosecond lasers with a maximum pressure in the 1 GPa range [15–19], but most often destructively in the nanosecond regime with pressures reaching tens of GPa [20–24]. Optical damage imposes severe restrictions including a very limited number of shots on a sample of small size or in a specialized environment such as a cryostat, poor shot-to-shot reproducibility for samples that are not uniform from one region to the next, and no possibility for many measurements on the same sample region in order to explore cumulative effects of repeated moderate shock loading.

In the following, we present a methodology for the excitation of non-destructive nonlinear SAWs in Nb:SnTiO₃ substrates, at high-repetition rate, limited only by the mechanical strength of the sample. Our technique is based on the spatio-temporal superposition of numerous laser-excited nanosecond strain waves for SAW amplification. Additionally, we present a femtosecond defocusing imaging technique that can adequately reveal with great sensitivity the nonlinear reshaping of the propagative SAW.

Various configurations enabling the spatio-temporal tailoring of laser sources for acoustic wave generation in the MHz frequency range have been explored in the past, ranging from a moving laser source [25], to an array of synchronized lasers [26], to the use of a White cell to split an input laser beam [27]. The current approach borrows the general idea while simplifying the required experimental apparatus. The set-up used in this study employs a free-space angular-chirp-enhanced delay (FACED) cavity [28], depicted in Fig. 1(a), whose purpose is to split an input pulsed laser beam into a train of sub-pulses spread in time and space. The device consists of two slightly angled high-reflective planar mirrors separated

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by a distance $d$. The input beam from an uncompressed Ti:Sapphire regenerative amplified laser (1 kHz repetition rate, 300 ps pulse duration, 800 nm central wavelength) is focused by a cylindrical lens near the back mirror of the cavity, propagates back and forth through the device, and reemerges at the entrance location. Each ray forming the input beam experiences a number of reflections that depends on its incidence angle. Consequently, the output of the device is a spatio-temporally spaced pulse train with an inter-pulse separation approximately equal to the round-trip time of the cavity, i.e., $\tau \approx 2d/c$, where $c$ is the light velocity. In this work, $d$ is fixed at about 30 cm, corresponding to an inter-pulse time separation $\tau$ of 2 ns.

The pulses exiting the FACED device are angularly dispersed, which enables each output beam to be cylindrically focused and directed to a separate location on the sample surface, as depicted schematically in Fig. 1(b). A time-integrated image of the excitation region is shown in Fig. 1(c). Each individual line gets absorbed in the material, inducing a thermoelastic response. This process launches acoustic waves that propagate in the vicinity of the surface, namely a surface acoustic wave (SAW) and a surface-skimming longitudinal wave (SSLW), moving at the Rayleigh wave velocity $v_R$ and longitudinal wave velocity $v_L$, respectively. By tuning the tilt angle $\alpha$ of the FACED cavity, the line spacing can be finely scanned until the spatio-temporal spread matches the propagation velocity of either wave. At specific angles $\alpha$, the coherent superposition of individual acoustic waves leads to the build-up of a large-amplitude wave in the phase-matched direction. The surface displacement $u_z$ induced by the passage of the acoustic waves is monitored through reflective interferometric probing [29] using two optical probes located on both sides of the excitation region. Figure 2 shows typical waveforms acquired from a niobium-doped strontium titanate (Nb:STO) sample at $1.0 \text{ J cm}^{-2}$ fluence. The waveform recorded for parameters tuned to amplify a SAW is shown in panel (a). The additive amplified wave is detected at the probe 1 location. Non-phase-matched waves, travelling in the opposite direction, are recorded by probe 2. Panel (b) shows the waveform recorded when amplifying a SSLW. Note that the surface wave selectivity can be obtained with minimal realignment of the optical set-up — by a simple mirror tilt.

In order to demonstrate the effectiveness of the acoustic generation platform, we performed experiments on phosphorus-doped silicon (P:Si, MTI Corporation) as well as Nb:STO (MTI Corporation). Si has been extensively studied in the past in the single-shot ablative shock regime [6, 7] and was used in this study as a benchmark for quantifying the SAW generation efficiency. STO is of interest due to the strain-induced ferroelectricity it exhibits starting from its normal paraelectric and low-temperature quantum paraelectric phases [30–32] as well as the effect of strain on its low-temperature superconductivity [11, 33, 34]. Above all, both Si and STO are among the most common substrates for sample deposition, and as such could be used in the study of strain-induced effects in materials that are either deposited as exfoliated 2D layers or grown as thin films on the substrates. A comparison of SAW generation in the current multi-line method to a single-line approach is shown in Fig. 3(a) for these two materials. For a fixed laser fluence, about an order of magnitude enhancement of surface displacements and corresponding strain amplitudes can be obtained on P:Si by the current generation method. For this sample, however, even by distributing the excitation energy over a large area, the maximum strains achievable are limited by the onset of laser-induced damage at the pump location. Still, it is noteworthy that the maximum strain amplitudes reached by the current technique without damage are very close to those achieved in a single-shot destructive manner in Si [7].

Much higher strain amplitudes could be reached on a
Nb:STO substrate. As shown on the right vertical scale of Fig. 3(a), strain amplitudes of 1.0 % can be attained at a 1 kHz repetition rate, with strains up to 3.0 % observed in the finite-shot regime — when the total number of shots is limited to several hundreds. These high amplitudes stem in part from the higher coefficient of thermal expansion of STO, which is about ten times higher than for Si, but also from a careful choice of Nb doping level to make the optical penetration depth match the SAW depth profile of the material. A penetration depth of about 15 μm for the 800-nm pump light was obtained from a 0.7 % Nb doping. For this sample, multi-line excitation in the finite-shot regime enables the onset of plasticity to be reached before any damage is induced from the pump laser light in the irradiated region. In this plastic regime, corresponding to the fluence range from 1.1 to 2.6 J cm−2 and corresponding SAW strains above 1.1 %, each laser shot induces dislocations and plastic deformations that alter the mechanical integrity of the sample. At those fluences, repetitive laser shots, in the range of several hundreds, lead to mechanical failure from cumulative fatigue. Fig. 3(b) shows a post-mortem confocal microscopy image of a Nb:STO surface taken following an experiment in the plastic regime, at a pump fluence of 1.1 J cm−2. The laser excitation sequence was interrupted after 3,000 shots, before the mechanical damage reached the excitation region. Fracture and dislocations in the substrate, along with regions of material ejection, can be seen a few hundreds of microns away from the non-damaged pump region, in the direction of propagation of the amplified SAWs. Note that in these finite-shot experiments, damage first appears at the farthest location from the excitation region, with subsequent shots inducing fracture closer to the pump, monitored in real time using a video camera (see Supplemental Material [35]).

The onset of plasticity and mechanical failure can be explained by analyzing the nonlinear evolution of the SAWs as they propagate. High-amplitude waves will develop a steepening shock front with increasing peak stress as they propagate due to the elastic nonlinearity of the medium [6, 36–38]. The nonlinear SAW reshaping can be revealed by techniques sensitive to a spatial derivative of the waveform, such as Schlieren imaging or the shadowgraph technique [39]. In this work, a simple transient reflectivity set-up with a slightly defocused imaging system has been used to probe the SAW temporal evolution. Fig. 4 shows a series of snapshots of a Nb:STO(111) surface taken in reflection using a sub-picosecond 400-nm probe as the illumination source. The direction of propagation was chosen to be [TT2] to maximize the elastic nonlinearity [8, 40]. The contrast ΔR/R in the transient reflectivity maps of Fig. 4 stems from two contributions, namely the photoelastic effect [41] and Fresnel diffraction from propagation of the optical field over the defocus length Δz. The latter effect has been analyzed before in the context of phonon-polariton imaging [42]. For small propagation lengths of the optical field, it is most sensitive to the Laplacian of the surface displacement (see Supplemental Material [35]):

$$\frac{\Delta R}{R} \propto -\frac{\lambda \Delta z}{2\pi} \nabla^2 u_z(x, y),$$

with λ the probe wavelength. The contribution of defocusing to the measured contrast can reach tens of percent, dominating over the photoelastic contribution, easily revealing the location and shape of the acoustic wave. The predominant effect of the Fresnel diffraction has been further confirmed experimentally from the fact that the reflectivity contrast switches sign when Δz is reversed.

A reshaping of the SAW profile is revealed from the evolution of the reflectivity data shown in Fig. 4 as the SAWs move away from their generation location. The data show a significant growth of the positive reflectivity peak and
FIG. 4. Time-resolved 400-nm reflectivity images for SAWs traveling along the [T12] direction of a Nb:STO(111) sample. An arrow points to the SAW location in each frame. The excitation region is outlined with a dashed box. Profiles extracted from a cut of the reflectivity maps along the horizontal dimension are shown on the right.

FIG. 5. Numerical modeling of the nonlinear evolution of a SAW traveling along the [T12] direction of Nb:STO(111). (a) Longitudinal strain $u_{x,x}$ at the surface, obtained for experimental conditions matching those of Fig. 4. The arrow shows the increase in peak strain as the wave propagates. (b) Simulated reflectivity profiles.

of the negative peak, initially absent at 0.0 ns (this time is taken as the time the amplified SAW leaves the excitation region). This nonlinear evolution is confirmed by numerical simulations obtained from propagating an experimental interferometric displacement waveform at delays matching those of Fig. 4 (see Supplemental Material [35]). Fig. 5(a) shows the evolution of the longitudinal strain at the surface, extracted numerically from the particle displacements. As the SAW propagates, the compressive component of the strain grows in amplitude. The projection of the stress along particular slip systems eventually reaches the yield stress of the material, which explains how fracture first appears significantly away from the excitation region when the pump fluence is set just above the onset of plasticity. Fig. 5(b) shows the second derivative of the surface displacement, qualitatively matching the evolution of the reflectivity profiles shown in Fig. 4, which further confirms that nonlinear dynamics are captured from time-resolved reflectivity measurements. Note that the simulation in Fig. 5 shows a sharper shock front than the results in Fig. 4 because of the limited spatial resolution of our optical measurements.

To summarize, this work establishes a versatile non-destructive technique for the generation of high-amplitude strain waves in solid substrates. Notably, Nb:STO substrates, commonly used for sample deposition, make it possible to reach strain amplitudes of more than 1.0% (up to 3.0% or 10 GPa in the finite-shot regime) before any type of damage — mechanical or laser-induced — occurs. The current technique enables transduction of high amplitude surface waves into samples of interests deposited on the substrates, and will thereby facilitate the study of dynamic strain effects in correlated materials and other types of samples sensitive to strain. Additionally, the high surface displacements reached using the current platform enable the use of a simple transient reflectivity imaging apparatus to track acoustic nonlinearities in materials or to simply image acoustic waves with great sensitivity. Extension of this work to bulk compressional waves will be possible with less strongly absorbing samples including STO with a lower Nb doping concentration for reduced absorption of 800-nm pump light. More uniform light absorption through tens of microns (e.g., optical penetration depth of roughly 100 μm) will produce a thermoelastic stress that will drive a primarily compressional shock, as we have seen for shock generation with single intense pulses in the single-shot regime [20].

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I. EXPERIMENTAL SET-UP

The full platform is illustrated in Fig S1.

A. Pump path

The pump laser is derived from the uncompressed output of a 1 kHz Ti:Sapphire regenerative amplifier (Coherent Astrella) with a central wavelength of 800 nm and pulse duration of 300 ps FWHM. Up to 1.6 mJ of pulse energy is used as input for the excitation path. A half-wave plate and polarizing beam splitter combination is used as a variable attenuator to control the pump energy.

B. Probe path

The probe beam is a continuous-wave frequency-doubled Nd:YAG (Coherent Verdi) laser operating at a central frequency of 532 nm with an average power lower than 10 mW. The probe first diffracts on a transmissive binary phase mask. The optical field at the phase mask location is then relayed to the back focal plane of the objective lens by a Keplerian telescope; a beam block is placed at the Fourier plane of the telescope to filter out all diffraction orders

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besides the $\pm 1$ orders. The probes are focused on the sample on each side of the excitation region. Their reflections are recombined interferometrically on the same phase mask, and the intensity of the signal beam is detected using a fast photodiode connected to a high-bandwidth oscilloscope. As the amplified surface waves propagate through probe 1, the sample surface displacement $u_z$ induces an optical path difference $\xi = 2u_z$ between both arms of the interferometer, which is translated into a change of intensity $I$ of the signal beam [1]:

$$I(\xi) = \langle I \rangle \left[ 1 - V \sin \left( \frac{2\pi \xi}{\lambda} + \phi_0 \right) \right],$$

where $\langle I \rangle$ is the average intensity, $V$ is the interferometric visibility, $\lambda$ is the probe wavelength, and $\phi_0 \in [-\pi, \pi]$ is the phase offset of the interferometer. Note that the interferometric range is limited to a relative surface displacement $u_z = \pm \lambda/8 = \pm 66.5$ nm at the highest sensitivity point ($\phi_0 = 0$). However, that range can be extended by tuning the initial phase offset of the interferometer, which is achieved by a horizontal translation of the phase mask.

### C. Transient reflectivity imaging path

In addition to the components illustrated in Fig. S1, transient reflectivity imaging capabilities can be easily added to the set-up. The compressed output of the Ti:Sapphire regenerative amplifier (Coherent Astrella), with a pulse duration of about 90 fs FWHM, is first frequency doubled to 400 nm in a barium beta borate (BBO) crystal. A set of variable-length fibers is then used to delay the beam propagation. The beam is finally used as an expanded illumination source on the sample, and its reflection is collected on the CMOS sensor in a stroboscopic fashion. The acquisition of each active frame (with the acoustic wave) is followed by the acquisition of a reference frame (without the acoustic wave).
Fig. S2. Acoustic resonances obtained on Nb:STO from scanning the line spacing $\Delta x$ of the excitation pattern ($k = 2\pi/\Delta x$). Dotted lines are guides to the eye.

Fig. S2 shows two resonances obtained from scanning the line spacing $\Delta x$ of the excitation pattern, corresponding to the selective amplification of a SAW and a SSLW. The Rayleigh velocity and longitudinal velocity of the waves can be extracted from the position of the peak maxima, in addition to an independent measurement of the inter-pulse delay. At resonance, the temporal full width at half maximum (FWHM) of a SSLW is smaller than the FWHM of a SAW, as evidenced from the higher amplitude of the SSLW resonance in Fig. S2; this stems from the fact that the longitudinal wave velocity is about twice the Rayleigh wave velocity.

III. EVOLUTION OF SAW-INDUCED MECHANICAL DAMAGE

In order to monitor the evolution of SAW-induced mechanical damage on a Nb:STO(100) sample, the sample surface is imaged with 400-nm light during the irradiation sequence. The laser fluence is set to 1.1 J cm$^{-2}$. The repetition rate is set to 100 Hz, and an image is acquired every 20 shots. Fig. S3 shows select snapshots acquired during such an experiment. The damaged region enters the field of view of the camera after 600 shots and reaches the excitation region after 3000 shots.

IV. PHASE CONTRAST ENHANCEMENT BY DEFOCUSING

The following derivation is based on [2]. Consider a phase object $\phi(x, y)$ at $z = 0$ illuminated by a monochromatic plane wave of wavelength $\lambda$. The field after the object is given by

$$u(x, y, 0) = e^{i\phi(x, y)}.$$  \hspace{1cm} (2)

It is assumed that $|\phi(x, y)| \ll 1$ at every point, such that $u(x, y, 0) \approx 1 + i\phi(x, y)$. If the phase object is a single sinusoidal function, that is, $\phi(x, y) = A \cos(k_0 x)$, its profile can be fully converted to an amplitude profile by a slight propagation of the field $u(x, y, z)$ due to Fresnel diffraction (this is the Talbot effect). For an arbitrary phase profile, each Fourier component gets diffracted, and the phase profile will be partially converted to an amplitude profile as the field propagates along $z$. For this case, the field at $z = 0$ can be decomposed into its Fourier components:

$$u(x, y, 0) = 1 + \frac{i}{(2\pi)^2} \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \hat{\phi}(k_x, k_y) e^{i(k_x x + k_y y)}. \hspace{1cm} (3)$$
Fig. S3. Snapshots of the SAW-induced damage as a function of the number of laser shots.

For each Fourier component, the Fresnel integral gives, in the paraxial limit, the evolution of the field along its propagation axis:

\[ u(x, y, z) = \frac{1}{i\lambda z} \int_{-\infty}^{\infty} dx' \int_{-\infty}^{\infty} dy' u(x', y', 0) e^{ik[|x-x'|^2+(y-y')^2]/2z}. \]  

(4)

In Fourier space,

\[ \hat{U}(k_x, k_y, z) = e^{-i[k_x^2+k_y^2]z/2k} \hat{U}(k_x, k_y, 0), \]  

(5)

with \( k = 2\pi/\lambda \).

If the field propagates for a short distance relative to all major Fourier components of the phase object, that is, \( z \ll k/ \max (k_x^2) \) and \( z \ll k/ \max (k_y^2) \), then Eq. 5 can be expanded in powers of \( z \) to yield

\[ \hat{U}(k_x, k_y, z) = \left[ 1 - \frac{i z}{2k} (k_x^2 + k_y^2) \right] \hat{U}(k_x, k_y, 0) + O(z^2) \]  

(6a)

\[ = U(k_x, k_y, 0) - \frac{i z}{2k} (k_x^2 + k_y^2) \left[ \delta(k_x)\delta(k_y) + i\hat{\phi}(k_x, k_y, 0) \right]. \]  

(6b)

Therefore,

\[ u(x, y, z) = u(x, y, 0) - \frac{z}{2k} \nabla^2 \phi(x, y, 0). \]  

(7)

The intensity of the propagating field at a distance \( z \) from the object is given by

\[ |u(x, y, z)|^2 = 1 - \frac{z}{k} \cos (\phi) \nabla^2 \phi(x, y, 0) + O(z^2) \]  

\[ \approx 1 - \frac{z\lambda}{2\pi} \nabla^2 \phi(x, y, 0). \]  

(8a)

(8b)
The contrast is given by

\[
\Delta I/I = \frac{|u(x,y,z)|^2 - |u(x,y,0)|^2}{|u(x,y,0)|^2}
\]

(9a)

\[
= -\frac{z\lambda}{2\pi} \nabla^2 \phi(x,y,0).
\]

(9b)

It is proportional to the Laplacian of the profile of the phase object and scales linearly with the propagation distance.

V. NUMERICAL SIMULATIONS

As a short SAW pulse propagates across a nonlinear elastic medium, its shape changes gradually due to the nonlinear distortion. Within the quadratic approximation and assuming the nonlinear distortion is slow (in the sense that the change on a distance comparable with the pulse width is small), the nonlinear evolution can be understood as an interaction among the Fourier components that results in generation of sum and differential frequencies. Quantitatively, this process is described by the evolution equation system derived in [3]:

\[
i \frac{d}{d\tau} B_n = n q_0 \left[ \sum_{0 < n' < n} F(n'/n) B_{n'} B_{n-n'} + 2 \sum_{0 < n' < n} (n/n') F^*(n/n') B_{n'} B_{n'-n}^* \right].
\]

(10)

Here, \( B_n \) is the \( n \)-th Fourier component of the strain of the propagating wave, \( \tau \) is the propagation distance, and the function \( F \) characterizes the rate of harmonics generation.

The experimental waveform registered at the location close to the source is expanded in the Fourier series and fed to the evolution system of equations as the initial condition. The system is integrated numerically over the propagation distance, and finally the inverse Fourier transform gives the predicted waveform. The kernel function \( F \) depends on the second- and third-order elastic constants of the solid, and on the geometry of the problem if the solid is anisotropic. Note that in certain geometries, \( F \) may become complex-valued and possess both real and imaginary parts. The second- and third-order elastic constants of the Nb:STO sample used in our numerical simulations were taken from [4, 5].

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