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ABSTRACT

Single-shot picosecond (ps) laser-induced delamination allows for the direct generation of suspended membranes from a continuous metallic film, offering a promising platform for the control of ultrafast magnetization dynamics driven by acoustic waves. Using the picosecond-ultrasonics method, we demonstrate that long-lived low-frequency acoustic waves can be optically excited in the delaminated cavities. At the same time, higher-frequency modes above 60 GHz exhibit a surprisingly fast damping, following a scaling law incompatible with the expected attenuation mediated by phonon-phonon scattering. Comparing measurements between delaminated cavities and a benchmark nickel film in contact with the substrate, we link our findings to structural modifications of the nickel crystal induced by the delamination process.

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Opto-acoustic properties of materials^{1,2} can be exploited in a broad range of applications, for instance, to modulate light intensity,^{3,4} control magnetization dynamics,^{5,6} gravitational wave detection and quantum opto-mechanics,⁷ as well as ultrafast and all-optical switching^{8,9} and ultrasound detection techniques¹⁰ for the characterization of nanoscopic features in materials.^{11–13} Furthermore, elastic strain engineering uses stress to boost material properties,¹⁴ including enhanced electron mobility in semiconductors for more efficient photovoltaic devices¹⁵ and transistors.¹⁶ In mechanical engineering, the pursuit of resonators with high quality factors is pivotal for a broad range of

applications, from ultralow noise opto-mechanical quantum technologies¹⁷ to energy-efficient magneto-acoustics in spintronic devices.^{18,19} Notably, coupling between acoustic and magnetic degrees of freedom in thin-film cavities can lead to a resonant enhancement of magneto-optical effects.²⁰ These perspectives motivate the search for optimized nano-resonators with lower damping and enhanced propagation of acoustic waves.

In practice, removing the substrate below a thin film can provide high-quality resonators, since the energy of the acoustic pulse is physically trapped within the film due to an impedance mismatch.^{21,22} In a

seminal work by Kim and Bigot,⁶ picosecond acoustic pulses trapped within a freestanding nickel film were used to control magnetization precession on ultrafast timescales. Despite this, opto-acoustic control of magnetization dynamics has not yet moved past the proof-of-concept stage, due to the complex methodologies needed to fabricate the freestanding film. For example, in Ref. 6, the freestanding film was realized by first growing a sacrificial salt layer on top of a SiO₂ substrate. A thin nickel film was then grown on top and separated from the substrate by dissolving the salt layer in water. The freestanding film was then stretched and clamped onto a holder with a center hole. A simpler methodology could help realize the technological potential of ultrafast magnetization control in practical applications²³.

In this context, laser delamination was recently proposed as a way to deterministically pattern cavities detached from the substrate at only a fraction of the usual time and fabrication costs.^{24,25} It was shown that, by carefully tuning the laser fluence, micrometric regions where the nickel film detaches from the substrates are created.²⁵ Once properly controlled, this process promises direct on-demand patterning of suspended membranes on areas down to the microscale and supporting a unique combination of multiple excitations (magnetic, optical, acoustic).²⁰ So far, existing works have focused on topographical characterization of nickel cavities fabricated via laser delamination (from now on referred to as “delaminated cavities”), employing optical interferometry and electronic microscopy techniques. Currently, an experimental study of the opto-acoustic response of delaminated nickel cavities is lacking. In the different scenario of Ref. 6, a surprisingly high attenuation of the acoustic pulses was observed in the

freestanding film compared to a benchmark sample that did not undergo the substrate removal procedure. In the context of this paper, the complex thermo-mechanical dynamics²⁴ leading to the re-solidification of the nickel crystal in proximity of the detached region might affect acoustic-pulse propagation in different ways after the fabrication of the delaminated cavities. Overall, alterations of the crystal-line structure and the interface quality could reshape the opto-acoustic response of our structures beyond the targeted reduction of damping directly associated with the absence of a substrate. Understanding the mechanisms governing the propagation of optically induced picosecond acoustic pulses in delaminated cavities, therefore, represents a necessary next step to assess possible integration in actual devices.

In this work, we characterize the opto-acoustic response of large ($>10\ \mu\text{m}$) micrometric bubbles of freestanding nickel films fabricated via laser delamination. The freestanding nickel cavities were fabricated using a pulsed laser focused on a 230 nm thick nickel thin film evaporated on top of a SiO₂ substrate. For a laser fluence slightly (a few percent) below the ablation threshold, it is possible to obtain the formation of closed delamination cavities²⁵ (more details on the fabrication can be found in the [supplementary material](#), Note 1). We benchmark our findings with the better-known scenario of a nickel thin film in contact with a SiO₂ substrate⁵ (from now on referred to as “pristine film”). Scanning electron microscope (SEM) images of both the pristine film and a typical delaminated cavity produced by our method are presented in [Figs. 1\(a\) and 1\(b\)](#) (details on sample preparation for SEM imaging can be found in the [supplementary material](#), Note 2). As shown in [Fig. 1\(b\)](#), the ps-laser delamination induces the

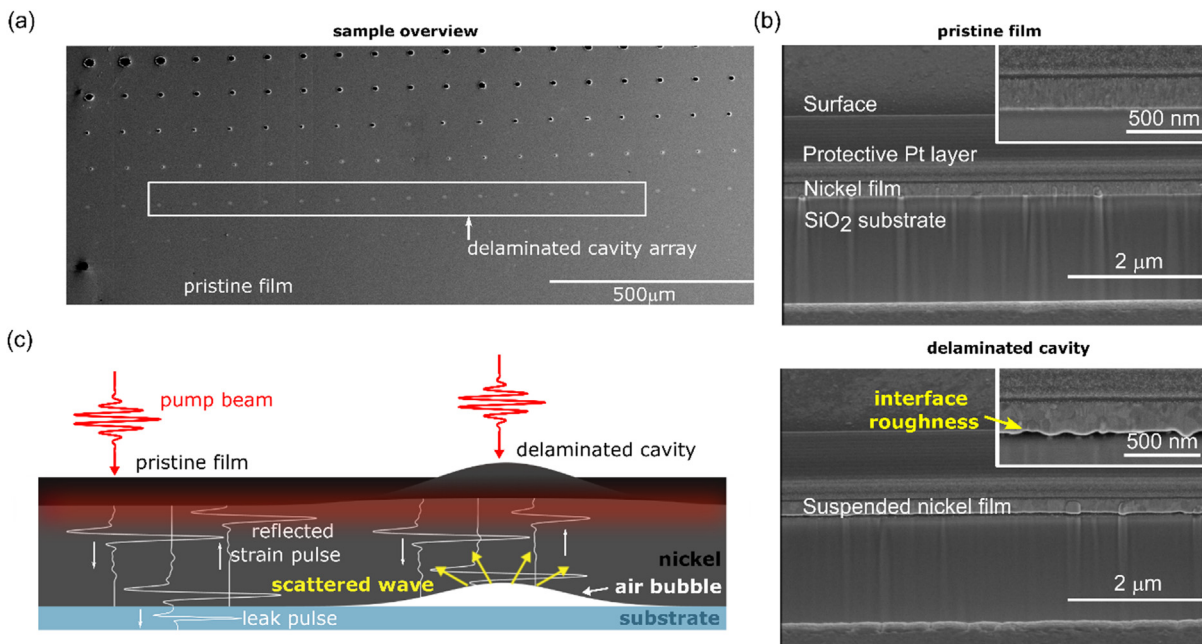


FIG. 1. (a) SEM overview of the patterned Ni film investigated in this work. The white rectangle highlights an array of delaminated cavities. For comparison, the circular structures in the top rows (black circles) are regions where the nickel is completely removed from the substrate (obtained for fluence values exceeding the ablation threshold). (b) SEM cross section cuts of the pristine nickel film (top) and delaminated cavity (bottom). Insets: magnified image of the pristine film and delaminated cavity cross section. The yellow arrow highlights the corrugations of the bottom nickel interface induced by the delamination process. The protective Pt layer was used only for imaging purposes and not in the cavity that was measured. (c) Sketch of the excitation of an acoustic pulse on the pristine film/delaminated cavity, after interaction with the pump pulse.

coalescence of small crystal grains of the pristine film into large crystallites in the delaminated cavity, accompanied by a significant increase in roughness at the nickel/air bottom interface.

We use the pump-probe method to investigate the propagation of picosecond acoustic pulses within the delaminated cavity. Comparing these findings with benchmark measurements on the pristine nickel film, we can understand how the ps-laser delamination process impacts the opto-acoustic performance of our structures. Additional information on the experimental setup can be found in the [supplementary material](#), Note 3. A sketch of the interaction between the pump beam and the sample is shown in [Fig. 1\(c\)](#). The pump beam, whose penetration is shorter (roughly 30 nm, corresponding to the optical skin depth of nickel at 515 nm) than the thickness of both the delaminated cavity and the pristine film, heats up the upper part of the system [shaded red region in [Fig. 1\(c\)](#)]. This localized heating triggers longitudinal acoustic waves that travel toward the lower part of the nickel domain. After reaching the lower nickel/air (delaminated cavity) or nickel/SiO₂ (pristine film) boundary, the acoustic pulse is partially reflected back to the first interface, where the probe pulse detects the effect of the strain on the transient optical response of the system. The periodic modulation of the refractive index due to the traveling acoustic wave shapes the pump-probe traces with a series of “echoes.” In the case of the film, we expect that the energy of the acoustic pulse is gradually dissipated due to transmission losses at the bottom nickel/SiO₂ interface. This effect should be significantly suppressed in the case of the delaminated cavity due to the higher impedance mismatch at the nickel/air bottom interface. However, the larger crystallites and higher roughness of the bottom nickel interface caused by the delamination process can enhance other loss mechanisms, for example, scattering of the acoustic pulse from defects.

Analyzing the pump-probe acoustic echoes in the time and Fourier domain, we can understand which mechanisms govern the time evolution of the acoustic pulse in the delaminated cavity. In [Fig. 2\(a\)](#), we show the transient reflectance curves measured under identical excitation conditions (fluence equal to 5 mJ/cm²) for five different delaminated cavities and two different spots on the pristine film. To better isolate the opto-acoustic response, an exponentially decaying baseline arising from electron–electron and electron–phonon scattering is subtracted from all pump-probe traces [see the [supplementary material](#), Fig. S4(a)]. The periodicity of the acoustic echoes depends on the thickness of the cavity/film and the sound velocity in nickel. Considering a thickness of 230 nm, and a sound velocity of longitudinal acoustic waves of 6.04 nm/ps,²⁶ we can expect a periodicity for the round trip of 76 ps, corresponding to a fundamental acoustic frequency of approximately 13 GHz. This number roughly matches the observed periodicity in the measured data for both pristine film and delaminated cavities [see [Fig. 2\(a\)](#)]. The shape of the acoustic echoes depends on the relative amplitudes and phases of the different acoustic eigenmodes propagating in the cavity. While the curves measured on different spots of the pristine film are practically identical [see the two top rows of [Fig. 2\(a\)](#)], we observe a different response between delaminated cavities, plausibly due to variations in the corrugation profile of the bottom interface.

Compared to the pristine film, we can, in general, appreciate a smaller attenuation of the acoustic pulse between successive roundtrips in the delaminated cavities, accompanied by a distortion of the acoustic echoes. For example, in delaminated cavity 3, the initial acoustic

pulse transforms into almost sinusoidal oscillations with a period of roughly 38 ps, indicating that after three roundtrips, the response is dominated by a mode at twice the fundamental acoustic frequency. These observations suggest that the various eigenmodes experience different reflection conditions at the bottom interface and that a mechanism more complex than a simple reduction of damping governs the overall opto-acoustic response of the delaminated cavities.

To better understand acoustic-wave propagation in our target and benchmark samples, in [Fig. 2\(b\)](#), we show a Fourier transform of the experimental transient reflectance curves. In all cases, we observe a series of peaks, each one corresponding to a different eigenmode contributing to the overall acoustic wave-packet propagating in the two systems. We can clearly distinguish seven peaks for both curves acquired on the pristine film. Interestingly, the fundamental eigenmode at 13 GHz does not yield the highest contribution and is significantly broader than the other peaks. A similar observation was also reported in the different context of picosecond ultrasonics on van der Waals nanolayers,²⁷ suggesting a particularly efficient dissipation of the fundamental eigenmode toward the substrate compared to higher order modes. Because of its larger wavelength, we can hypothesize that the fundamental mode experiences a higher degree of elastic coupling between the nickel and SiO₂,²⁸ leading to more efficient dissipation toward the substrate. As a result, the opto-acoustic response is mainly determined by the $n = 2, 3, 4$ order eigenmodes, with the dominance of the 3rd mode at ~40 GHz. On the other hand, the fundamental mode is clearly contributing to the spectra of the delaminated cavities, while all eigenmodes above 80 GHz are completely suppressed. Overall, the acoustic spectrum of the response of the cavities is dominated by lower frequency modes compared to the film. This analysis is another way to see the different “broadness” and shape of the strain profile in the cavity compared to the film. The spectral position of these eigenmodes can be estimated by following a simple approach. The delaminated cavity can be modeled as a homogeneous nickel layer of thickness h with free external faces, and due to the symmetry of the excitation, only the longitudinal modes can be excited. The eigenfrequencies satisfy the following relation:²⁹

$$f_n = \frac{v_l}{2h} n, \quad n = 1, 2, 3, \dots, \quad (1)$$

where v_l is the longitudinal speed of sound. As for the film, it can be modeled as a nickel layer on top of a semi-infinite SiO₂ substrate. The nickel layer leaks out mechanical energy to the substrate; hence, the eigenfrequencies f are complex, their imaginary part representing the acoustic damping. Since the acoustic impedance of nickel ($Z_{\text{Ni}} = 5.37 \text{ kg m}^{-2} \text{ s}^{-1}$) exceeds the one of the SiO₂ ($Z_{\text{SiO}_2} = 1.31 \text{ kg m}^{-2} \text{ s}^{-1}$), the real part of the eigenfrequencies obeys Eq. (1) as well.³⁰ The position of the eigenfrequencies is highlighted in [Figs. 2\(c\) and 2\(d\)](#) with blue (film on top of the SiO₂ substrate) and red (freestanding film) triangles. The eigenfrequencies estimated from Eq. (1) agree well with our experimental findings, confirming that the peaks shown in the [Fig. 2\(b\)](#) correspond indeed to longitudinal acoustic modes.

To further support our discussion, we developed an opto-thermo-mechanical model and solved it with the finite element method (FEM) implemented in Comsol Multiphysics® (see the [supplementary material](#), Note 4, for further details).^{31–33} The pump pulse at 515 nm illuminates from the nickel layer from the top (we

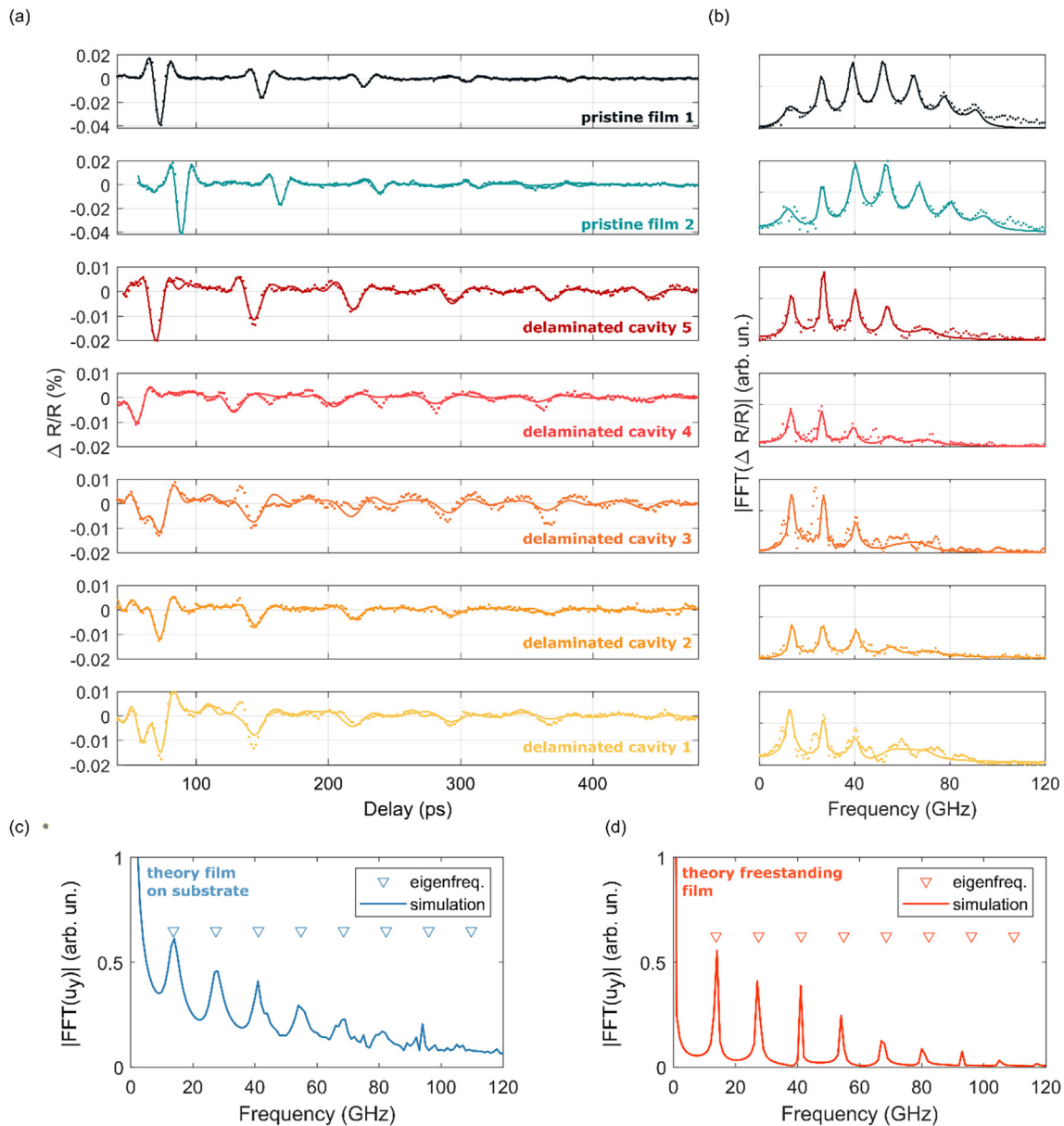


FIG. 2. (a) Transient reflectivity measurements (dots) and fit (solid line) for two spots on the pristine nickel film and five different delaminated cavities. (b) Fast-Fourier-transform of the curves in (a). (c) Spectral position of the expected eigenfrequencies (triangles) and FFT of the simulated displacement field u_y for a thin film on top of a SiO_2 substrate with thickness identical to the nominal value of our samples. (d) Same as (c), but for a freestanding film.

assume a fluence equal to 5 mJ/cm^2 , as in the experiments) and penetrates for less than 30 nm (see the [supplementary material](#), Fig. S5). This causes a localized increase in electronic temperature in the nickel layer close to the air side up to 800 K, just after the pump pulse has interacted with the system (see also the [supplementary material](#), Fig. S6). The electrons transfer energy to the phonon population, and hence, a phononic temperature increase occurs. On the air side of the Ni layer, the phononic temperature reaches 150 K approximately 2 ps after pump excitation (see the [supplementary material](#), Fig. S6). After

2 ps, a thermal expansion of the first 30 nm of the nickel layer takes place, triggering the generation of an acoustic pulse, which travels toward the backside of the Ni layer and is reflected when it arrives at the second interface. The reflection is only partial in the case of the film on top of the substrate, whereas the totality of the mechanical energy is reflected, in principle, at the bottom boundary of the freestanding film. This can be appreciated in the simulations where for the freestanding we have an almost infinite number of echoes, while the echoes vanish quickly for the film on top of the SiO_2 substrate (see the

supplementary material, Figs. S7 and S8, respectively). After the reflection, a wavefront propagates toward the top part of the nickel layer and when it arrives in the first 30 nm of the layer—the zone interacting with the probe beam—a transient variation of the mechanical displacement (see the supplementary material, Note 4) due to the echo is recorded by the probe, changing the transient reflectance signal detected in our setup. The FFT transform of the longitudinal displacement u_y calculated with FEM is reported in Figs. 2(c) and 2(d) (continuous lines), showing good qualitative agreement with the peaks observed in the FFT of the transient reflectivity traces and confirming the consistency of the model with the experimental data and observations. The minor discrepancies between the calculated eigenmodes and the experimental ones can be attributed to the idealized conditions assumed in our simulations, which are performed on an ideal system. In contrast with the experimental results, it is also worth mentioning that, in our simulations, the film shows an intense peak at the fundamental frequency. This discrepancy can also be explained by assuming that the experimental curves are affected by zero detection efficiencies at certain frequencies, resulting from the zero-overlap integral between the acoustic eigenmode and the photo-acoustic sensitivity function.³⁴ Remarkably, the comparison between experimental and theoretical curves unambiguously shows that our simple model, where the delaminated cavity is treated as an ideal freestanding film, cannot fully

capture the physics governing propagation of acoustic pulses. Grain boundaries and surface irregularities play an important role in shaping the opto-acoustic response of the delaminated cavities outside the simple reduction of attenuation.

Disentangling the dynamics associated with individual eigenmodes can provide valuable insight into our experimental findings. To achieve this, we calculate a short-time Fourier transform (STFT) for all measured pump-probe traces. The resulting spectrograms are shown in Fig. 3. The two spectrograms associated with the pristine film [Figs. 3(a) and 3(b)] reveal how the fundamental eigenmode is barely excited, and its contribution vanishes completely after the first round trip. The excitation of the second eigenmode is also comparatively weak, while modes $n = 3, 4, 5$ dominate the response. Regardless of the initial excitation amplitude, all modes in the pristine film undergo significant attenuation above 50% after each round trip. In contrast, the first two eigenmodes are efficiently excited in the delaminated cavities, and their lifetime is considerably longer compared to the pristine film. Remarkably, in two of the cavities considered (cavities 3 and 4), the $n = 2$ mode at 26 GHz dominates the response and propagates essentially unperturbed over the time window we investigated. In agreement with our previous considerations, the spectrograms further highlight how the acoustic wave-packet propagating in the delaminated cavities lacks high-frequency modes above 60 GHz.

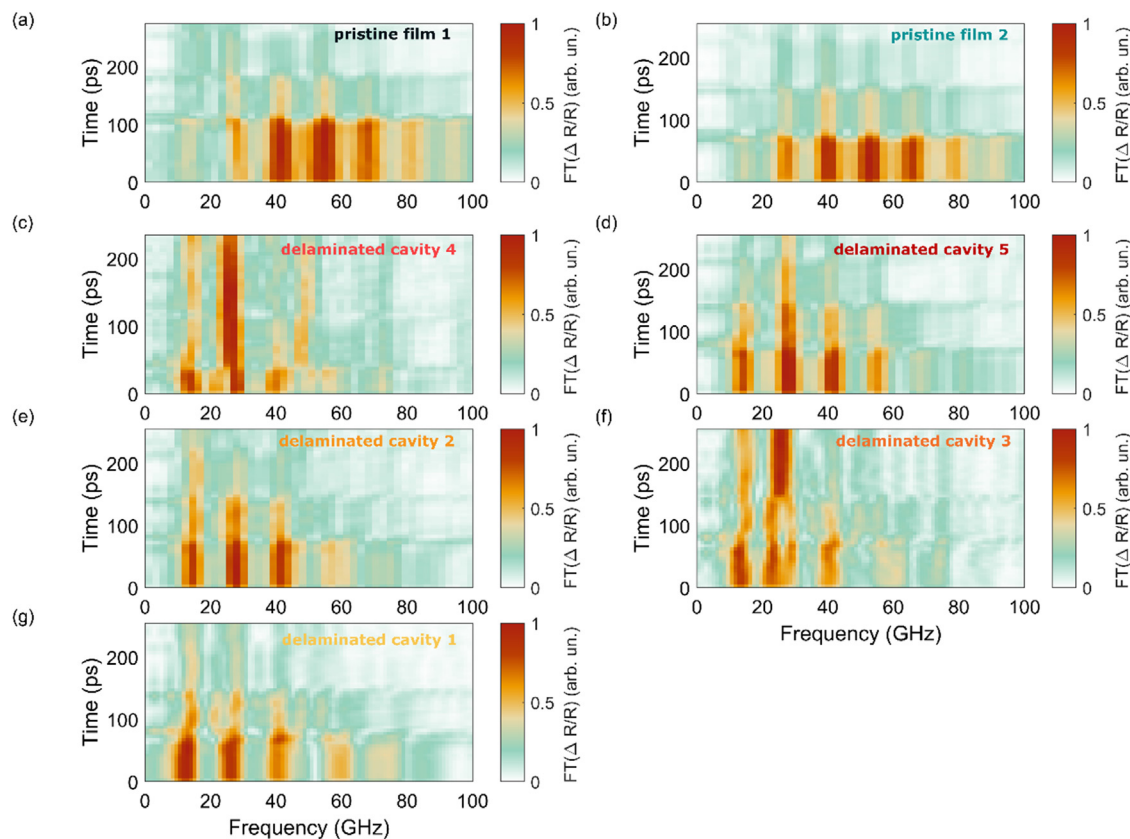


FIG. 3. Propagation of individual acoustic eigenmodes for the pristine film (a) and (b) and for the different delaminated cavities (c)–(g). The spectrograms are obtained from the experimental pump-probe traces shown in Fig. 2(a) using a short-time Fourier transform algorithm.

Comparison of the time evolution of the individual eigenmodes suggests that acoustic-pulse propagation in the pristine film and delaminated cavities is governed by different mechanisms.

To understand the dominant process in the two cases, we evaluate the damping associated with each eigenmode. To achieve this, we decompose the differential reflectance curves in a series of damped harmonic oscillators, each corresponding to an acoustic eigenmode of the pristine film/delaminated cavity. In particular, we fit our experimental differential reflectance pump-probe traces with the function

$$\frac{\Delta R(t)}{R} = \sum_{n=1}^N A_n e^{-\gamma_n t} \cos(\omega_n t + \phi_n), \quad (2)$$

where we consider N damped oscillators with amplitude A_n , decay rate γ_n , and eigenfrequencies $f_n = \omega_n/2\pi$. As indicated by the Fourier analysis discussed above, respectively, $N=5$ and $N=7$ eigenmodes are needed to reproduce the response of the delaminated cavities and pristine film. The good agreement between the experimental data and best fit curves, both in the time and frequency [solid curves in Figs. 2(a) and 2(b)] domain validates the soundness of our methodology.

In Fig. 4(a), we plot the extracted decay rates γ_n as function of frequency f_n for the eigenmodes considered. The error bars are obtained from the fitting procedure.

In Ref. 24, Ghita *et al.* applied the same analysis to the pump-probe measurements acquired by Kim *et al.*⁶ on a freestanding film

obtained via dissolution in water. A quadratic dependence of the decay rate on frequency was observed up to 80 GHz, indicating that that phonon-phonon scattering is the main attenuation mechanism. In our case, a fit of the decay rates to a power law $\gamma = a + bx^c$ yields for both spots investigated on the pristine film an exponent $c \simeq 1.5$, in good agreement with the observations of Ghita *et al.* On the other hand, the measurements on the delaminated cavities yield a strikingly different $c = (4.6 \pm 0.8)$. This faster-than-quadratic dependence demonstrates that the damping of acoustic waves in the delaminated cavities is governed by a mechanism other than phonon-phonon scattering. On the other hand, a $\gamma \propto \omega^4$ dependence is expected when attenuation is dominated by Rayleigh scattering.^{35,36} In this context, the $c \simeq 4$ exponent strongly indicates that scattering due sub-acoustic-wavelength irregularities induced by the fabrication process [such as the larger crystallites and the irregularities at the bottom interface visible in the SEM images of Fig. 1(b)] governs the high-frequency opto-acoustic response of the delaminated cavities.

Finally, in panel 4(b), we extract the quality factors $Q_n = \omega_n/2\gamma_n$ associated with the various eigenmodes and compare them with the typical quality factor $Q_G \simeq 12$ of magnon resonances in nickel.²⁴ As discussed by Ghita *et al.*, the condition $Q_n \geq Q_G$ is a necessary requirement for efficient excitation of magnetization dynamics by the acoustic pulse. This condition is satisfied by the second and third eigenmodes in all cavities, demonstrating that delaminated cavities can host acoustic pulses suitable for control of magnetization dynamics³⁷

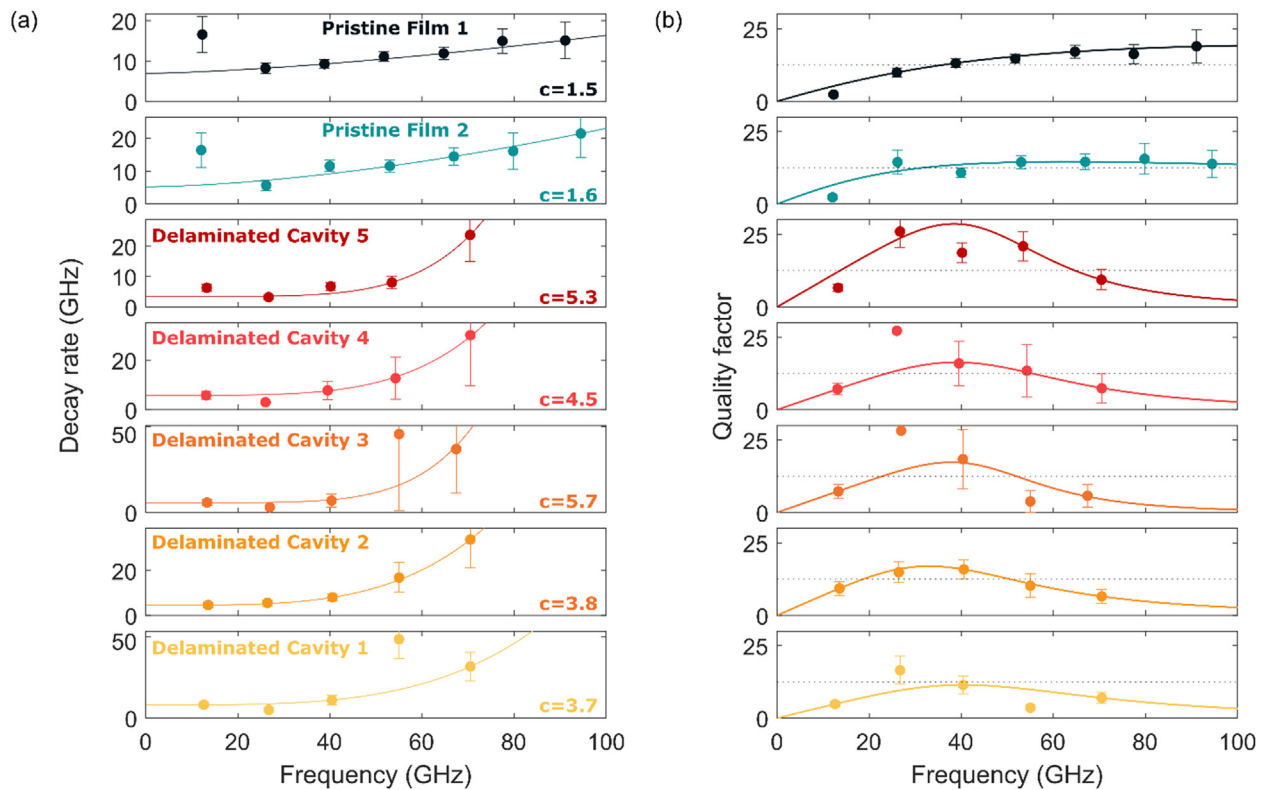


FIG. 4. (a) Decay rates as a function of frequency obtained from the fit of the pump-probe data. A power law fit $\gamma = a + bx^c$ is shown as a solid line as a guide to the eye for all cases. (b) Q-factor associated with different eigenmodes. The solid lines are obtained from the fits of panel (a) and shown as a guide to the eye. The black dashed line indicates the Q-factor of magnon resonances in nickel $Q_G \simeq 12$ as discussed in Ref. 20.

up to 60 GHz. Above this frequency, we observed a decrease in quality factor. The frequency-scaling points toward phonon-defect scattering as the main attenuation mechanism, a behavior we attribute to structural changes in the nickel layer induced by the delamination process.

In conclusion, we performed an ultrafast spectroscopy study of acoustic phonons excited by femtosecond light pulses and confined in a cavity of a freestanding nickel layer fabricated using laser delamination. Combining a Fourier analysis with numerical multi-physics simulations, we showed that the freestanding cavity detached from the substrate displays a different acoustic response, with a significantly weaker damping of low-frequency modes compared to a reference thin film on a SiO₂ substrate. However, we also observed a surprisingly efficient suppression of higher-order modes in the delaminated cavities. To understand the origin of this behavior, we quantitatively estimated the damping associated with the different eigenmodes of both the delaminated cavities and the pristine film. While the damping of the pristine film is compatible with the expected quadratic frequency-dependence, a fourth-order power law best describes the observations on the delaminated cavities. These findings, supported by SEM images, show that the delamination process causes the formation of defects in the nickel crystal, which ultimately limit the high-frequency response of freestanding cavities. Finally, we estimated the quality-factor associated with the different eigenmodes and compared it with that of the magnon resonances of nickel. Our analysis shows that delaminated cavities are indeed a promising platform for ultrafast modulation of acoustic waves. We showed that opto-acoustic control of low-frequency (<60 GHz) magnetic excitations is already viable. While high-frequency (>100 GHz) operation remains a desirable goal for technological applications, our findings indicate that excitation of >60 GHz acoustic modes is currently inefficient in delaminated cavities. Crucially, we identified scattering between phonons and fabrication-induced defects as the limiting factor. In view of these findings, we envision that further optimization of the fabrication technique can overcome current limitations, paving the way for faster modulation of acoustic pulses in delaminated cavities.

See the [supplementary material](#) for details on fabrication of nickel cavities, sample preparation for SEM imaging, pump-probe measurements, numerical simulations, and Fourier analysis of the single echoes.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Alba Viejo Rodríguez, Andrea Rossetti, and Marco Gandolfi contributed equally to this work.

Alba Viejo Rodríguez: Data curation (equal); Formal analysis (equal); Investigation (lead); Methodology (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Andrea Rossetti:** Data curation (lead); Formal analysis (lead); Investigation (equal); Methodology (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Marco Gandolfi:** Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Yoav Urbina Elgueta:** Formal analysis (supporting); Investigation (supporting). **Evgeny B. Modin:** Formal analysis (supporting); Investigation (supporting). **Svetlana Starikovskaia:** Investigation (supporting). **Tat Loon Chng:** Investigation (supporting). **Vasily Temnov:** Investigation (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (supporting). **Maria Antonietta Vincenti:** Formal analysis (supporting); Investigation (supporting); Methodology (supporting); Validation (supporting); Writing – review & editing (supporting). **Daniele Brida:** Investigation (supporting); Methodology (supporting); Resources (supporting); Validation (supporting); Writing – review & editing (supporting). **Paolo Vavassori:** Conceptualization (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Nicolò Maccaferri:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (lead); Resources (lead); Supervision (lead); Validation (equal); Visualization (equal); Writing – original draft (lead); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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