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Enabling resonant ultrasound spectroscopy in high magnetic fields

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ABSTRACT:

Resonant ultrasound spectroscopy (RUS) is a powerful method to determine elastic constants with high accuracy and precision from a single measurement of the mechanical resonances of a sample. Conventionally, the quantitative extraction of elastic moduli with RUS assumes free boundary conditions which can often lead to the adoption of unstable sample positioning between ultrasonic transducers that is incompatible with extreme environments like high magnetic fields. We show that, under specific conditions, introducing a small amount of adhesive between a RUS sample and ultrasonic transducers introduces a perturbation to the free resonance condition which can be accounted for by a simple model. This means elastic constants can be determined to within the uncertainty of conventional RUS, but with significant improvements including sample stability and control of sample orientation. We demonstrate the efficacy of this approach with measurements on a range of materials including room temperature measurements on polycrystalline metals, temperature-dependent measurements of the structural phase transition in strontium titanate single crystals, and magnetic field-dependent measurements of magnetic phase transitions in gadolinium polycrystals up to 14 T. © 2024 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

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I. INTRODUCTION

Elastic constants are second derivatives of the free energy with respect to strain which describe the resistance of a material to elastic deformation. They are indispensable in characterizing mechanical performance^{1–3} and sensitive to atomic structure and bonding⁴ in all types of materials.⁵ As the thermodynamic susceptibility of strain, elastic constants also reflect strain couplings to structural, electronic, and magnetic degrees of freedom.^{6–8} This makes elastic constants ideal to understand changes in materials as a function of external fields (temperature, magnetic field, etc.) and symmetry-breaking at phase transitions.^{6–8}

There are numerous static, quasi-static, and dynamic methods to measure elastic constants.⁹ Among these, resonant ultrasound spectroscopy (RUS)¹⁰ is a particularly powerful technique for the non-destructive examination of elastic properties because the mechanical resonances of a sample with a known geometry and mass provide access to the complete set of elastic constants, as well as symmetry-resolved ultrasonic attenuation,^{9,11} with high accuracy and precision.^{12–14} The simultaneous determination of the entire elastic constant tensor makes RUS well-suited for studying elastic anisotropy (e.g., texture in metals¹⁵) and the level of precision enables detailed studies of phase transitions with relatively small elastic signatures [e.g., superconducting and skyrmion phase transitions correspond to relative changes in

elastic constants $\sim 10^{-5}$ – 10^{-3} (Refs. 16–18)]. Together, these aspects have made RUS an important tool for studying elastic properties with implications in both fundamental and applied science.

Significant progress has been made since the inception of RUS,^{19–22} especially in the areas of computational efficiency²³ and instrumentation.^{24,25} Recent advances in generalizing RUS to complex geometries through finite-element²⁶ or surface-mesh²⁷ approaches also offer promising paths toward efficient elastic constant determination on irregularly shaped samples. Central to all these approaches is that information about elastic moduli is encoded into mechanical resonant frequencies, with the exact relationship between the two depending on geometry, materials properties (density and internal symmetry), and mechanical boundary conditions. Free boundary conditions are most often employed in RUS inversion because they yield well-defined solutions and make the inversion computationally efficient.^{12,13,28}

While free boundary conditions are most commonly used to quantitatively extract elastic moduli from measured resonant frequencies, RUS experimental implementations only approximate free boundary conditions. The most prevalent RUS methods require transducers to contact the sample and the resulting contact forces cause deviations from free boundary conditions which shift resonant frequencies from their free resonance value.²⁹ In well-designed experiments, these forces are sufficiently small and applied at an appropriate location on the sample such that the resonant frequency shifts are below the uncertainty of the inversion

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process.¹³ Contact effects are a negligible perturbation to the free resonance condition in these scenarios, but computational simplicity comes at the expense of sample stability: samples are typically positioned in unstable configurations to minimize contact with transducers. Therefore, RUS is ill-suited for experiments in “extreme” environments where sample stability is paramount, such as high magnetic fields. RUS approaches which utilize non-contact ultrasonic excitation/detection³⁰ offer closer approximations to the free resonance condition than transducer-based measurements, but are only compatible with electrically conductive materials,^{31,32} necessitate coating samples with thin films,³³ or face similar sample stability challenges as transducer-based RUS owing to the need to support samples.³⁴ Additionally, while sample orientation is often not considered in conventional RUS, orientation becomes important if an external field (magnetic,^{17,18,35,36} electric,³⁷ etc.) must be applied along a specific direction in a sample. While there have been some efforts towards applying RUS in magnetic fields,^{17,18,35,36} they involved immense work to maintain sample positioning, are incompatible with samples with appreciable magnetic moments, and/or are limited to low magnetic fields. Together, sample movement and orientation represent two significant challenges which limit practical applications of RUS in extreme environments.

The central result of this paper is that the use of adhesives between samples and ultrasonic transducers, under specific conditions, introduces a perturbation to the free resonance condition akin to the effect of contact in conventional RUS. The resonant frequency spectrum obtained in this manner is similar to conventional spectra measured under free boundary conditions with a shift in the resonant frequencies to higher values. These effects can be captured by a simple model and effectively minimized, such that elastic constants are determined when a sample is adhered to transducers to within the same uncertainty of conventional RUS, but with the benefits of enhanced sample stability and controlled sample orientation imbued by the adhesive. Moreover, efficient elastic constant analysis is maintained

because established RUS-inversion frameworks are compatible with this approach. After showing that we recover the accuracy of conventional RUS, we demonstrate these adaptations enable RUS measurements of changes in elastic constants and ultrasonic attenuation in magnetic fields up to 14 T with a comparable sensitivity to phase transitions. This advancement makes RUS amenable to high magnetic fields, and other extreme environments, with improved reproducibility, sample stability, and control over sample orientation.

II. METHODS

RUS measurements were performed on a custom-built system using a similar experimental approach described in Ref. 24 and summarized in Fig. 1. A sample shaped into a suitable geometry for RUS (rectangular parallelepiped or cylinder) was placed between two ultrasonic transducers. The transducers consisted of a metallic enclosure filled with an epoxy, a piezoelectric active element (lead zirconate titanate for ambient measurements and lithium niobate for low temperature measurements), and an alumina or sapphire hemisphere (to electrically insulate the piezoelectric from conductive samples, minimize wear on the piezoelectric, and guarantee point contact with the sample³⁸). For conventional RUS, care was taken to ensure the sample was lightly held between the transducers and the sample was positioned to maximize the number of measurable resonances. Then, a field-programmable gate array-based system generated a single-frequency excitation in one transducer and measured the response to this excitation frequency with the other transducer.²⁴ Sweeping the excitation frequency allowed for the identification of the resonant frequencies of the sample from modified Lorentzian fits to the frequency spectrum. For the geometries used in this work, elastic constants were then obtained from measured resonant frequency spectra via an iterative, Lagrangian minimization approach.²³ Temperature-dependent measurements were performed on a custom-built piston probe^{17,18} and a custom-built cantilever probe compatible with a Physical Property Measurement System (Quantum Design).

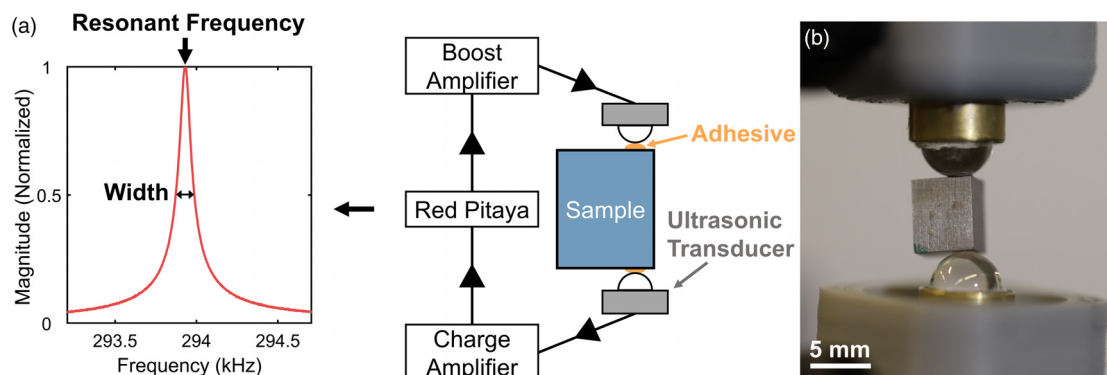


FIG. 1. (Color online) (a) Overview of resonant ultrasound spectroscopy experiments. A sample placed between two ultrasonic transducers is subjected to a single-frequency ultrasonic excitation from one transducer, and the response to this excitation is measured by the other transducer. Sweeping the frequency yields a resonance spectrum where each resonance is characterized by a resonant frequency and width. For conventional RUS, the sample is lightly held between the two transducers. Here, we adhere the sample to each transducer at a point. (b) Picture of a RUS experiment where adhesive was used to hold the sample (a high-density tungsten alloy) to the transducers.

The data reported here were obtained on four samples. Data at ambient conditions were collected on 1100 aluminum alloy³⁹ and high-density tungsten alloy (90% W, 7% Ni, 3% Fe) samples.^{15,40} Temperature-dependent measurements on a single crystal SrTiO₃ sample (Appendix C) and temperature- and magnetic field-dependent measurements on a pure gadolinium polycrystal were used to examine the sensitivity to well-known phase transitions.^{41,42} These four samples were selected because they span a range of geometries, masses, stiffnesses, and elastic-anisotropies. The approach has also been successful in additional polycrystalline and single crystal systems which will be the subject of future work. The adhesive used here was a commercially available cyanoacrylate-based adhesive which could be removed from the samples with acetone. Other fast curing adhesives were also tested, yielding comparable results to those presented in this work. The amount of adhesive used for optimized adhesion was ≤ 0.5 mg for samples with masses and dimensions spanning 0.1–2 g and 3–13.5 mm, respectively.

III. RESULTS

First, we consider the effects of coupling the sample and transducers via an adhesive by measuring the resonance spectra of an 1100 aluminum alloy sample without an adhesive, with a small amount (≈ 2 mg) of adhesive, and with a large amount (≈ 20 mg) of adhesive (Fig. 2). When the sample is adhered to the transducers, all the resonant frequencies of the sample are shifted to higher frequencies and broadened, and the amplitude of the resonances is increased. Figure 2 demonstrates that these effects become more pronounced as the coupling between the sample and transducer is increased by using more adhesive. As discussed later, the increased amplitude may have important applications in non-linear RUS measurements or for poorly resonating materials like polymers or geological materials.^{43,44} However, for the purposes of elastic constant determination, frequency shifts and broadening are deleterious: shifts in the resonant frequency will affect elastic constant determination from the inversion process, and excessively broadened

peaks can be difficult to identify leading to missing resonant frequencies.⁴⁵ For example, the individual resonances in the frequency range of 280–320 kHz become difficult to distinguish when an excessive amount of adhesive is used [top-most spectrum in Fig. 2(a)]. In such cases, the elastic constants will not be able to be fit from the resonance spectrum, or they can be fit but will be erroneous.

Figure 2 suggests that if the coupling between the sample and transducers is sufficiently small (i.e., the amount of adhesive used is similar to or less than amount used to obtain the middle spectrum in Fig. 2), the coupling effects may be treated as a perturbation to the free boundary condition. This approach is akin to how one can perturbatively treat geometric imperfections.⁴⁴ To develop a semi-quantitative understanding of the effects of coupling the transducers and sample with an adhesive, we assume the adhesive acts as a linear anelastic body, forming a negligible contact area between the transducer and the sample. Then, as derived in Appendix A, the resonant frequency of this coupled sample-transducer mechanical system, f' , is related to the free resonant frequency of the sample, f_0 , by

$$f'^2 = f_0^2 + \frac{m_g}{m_s} \frac{Y_g}{4\pi^2 \rho_g t_g^2}, \quad (1)$$

where m_s is the mass of the sample and Y_g , ρ_g , m_g , and t_g are the Young's modulus, density, mass, and thickness of the adhesive, respectively. Equation (1) indicates the principal effect of the adhesive is to shift the free resonant frequencies (f_0) of the sample to higher values (f') by a constant that is related to the properties of the adhesive and the ratio of the adhesive mass to the sample mass. While in our simplified treatment each resonant frequency has the same shift, it is likely that the shift is different for each resonant frequency. Regardless, since the shift is a constant, changes in resonant frequencies (and elastic constants) will be unaffected by the adhesive. This has important implications for studying temperature- and magnetic field-dependences of elastic constants with RUS where resonant frequency changes are the relevant measured quantity.⁴⁶ Furthermore, Eq. (1) suggests the shift can be minimized by

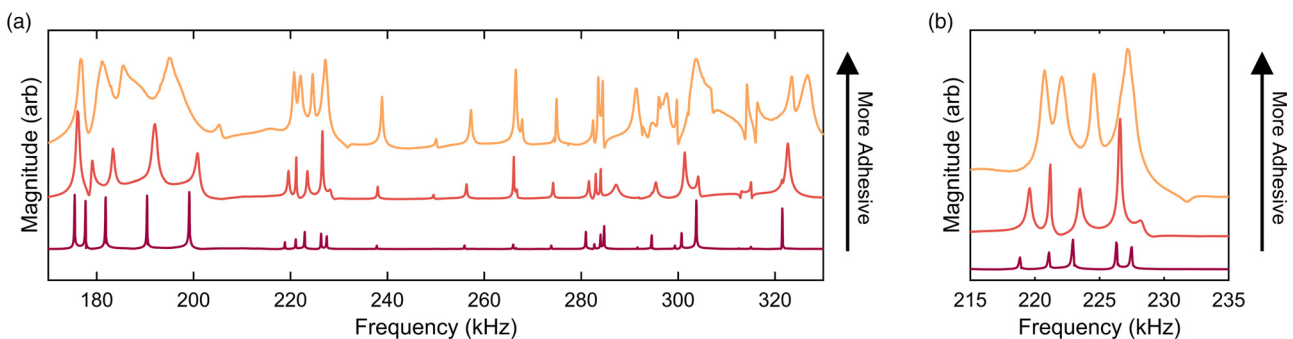


FIG. 2. (Color online) (a) Resonant ultrasound spectra acquired on the same sample (Al 1100 rectangular parallelepiped) with different amounts of adhesive. (b) Zoom of the spectra in (a). Coupling the sample to the transducers with an adhesive increases the amplitude of the resonances and causes the resonant frequencies of the sample to be shifted to higher values and broadened. Stronger coupling (more adhesive) amplifies these effects. Spectra have been shifted vertically for clarity.

utilizing relatively massive samples, choosing softer adhesives, or reducing the amount of adhesive. As the sample size is often unchangeable and we are interested in using stiff adhesives to keep samples from moving in extreme environments, we focus on minimizing the amount of adhesive.

In addition, it may be important to consider the location at which the adhesive is applied to the sample. In conventional RUS, the point of contact between the sample and the transducers can affect elastic constant determination because certain resonances may not have measurable amplitudes where the transducer contacts the sample or may be strongly affected by the transducer weight. Given the complex shapes of the resonant modes, the ideal location to apply the adhesive may differ for each sample. In principle, the resonant mode shapes can be measured or simulated (e.g., Refs. 26, 47) for a particular experiment. In practice, adhering the sample close to its corners appears to be sufficient. This works particularly well in our experiments because the hemispheres on the transducers inherently minimize the contact area of the adhesive.

Figure 3 demonstrates that when only a small, optimized amount of adhesive is used (≤ 0.5 mg), sharp resonances from the sample are present which exhibit small frequency shifts and increased width. The resonances measured from the adhered sample form circles when plotted in the complex plane indicating they possess Lorentzian shapes, just like those measured with conventional RUS [Figs. 3(b) and 3(d)]. To obtain the same amplitudes in resonance spectra with and without the adhesive, the excitation voltage used with the adhesive was reduced by a factor of ~ 3 because of the enhanced coupling between the transducers and the sample. A consequence of this stronger

coupling is that direct transmission between the transducers is possible and spurious resonances from the system may be measured. Care should be taken to minimize these effects by reducing the excitation voltage.

To quantify the changes in the resonance spectra stemming from the adhesive, we compare the resonant frequencies obtained on a high-density tungsten alloy sample with and without an adhesive (data in Fig. 3 and Tables II–IV in Appendix B). Figure 4 demonstrates that the change in the frequencies caused by the adhesive is well-described by Eq. (1), i.e., the dominant effect of the adhesive is a shift in the resonant frequencies of the sample to higher values. The slope of this line is 1 within the uncertainty of the fit and the intercept (which is a measure of the size of the adhesive effect) is small compared to f_0^2 . Moreover, fixing the slope to 1 in Fig. 4(a) has a minimal impact on the fit quality or the elastic constant determination described below. A direct consequence of Eq. (1) is that higher resonant frequencies possess smaller relative changes from the adhesive. At sufficiently high frequencies (resonance number ~ 15 in this case), these variations are within the usual reproducibility of conventional RUS as determined by changing transducer positioning on the sample. This suggests that it may be possible to perform quantitative RUS measurements of elastic constants without the corrections described below if only higher frequency resonances can be used. Last, we find no systematic relation between the frequency shifts and the composition or symmetry of the resonance. This supports using the simple model in Eq. (1), which does not include mode-dependent corrections.

Having established the frequency shift stemming from the use of an adhesive, we now demonstrate that elastic constants determined with an adhesive are within the uncertainty

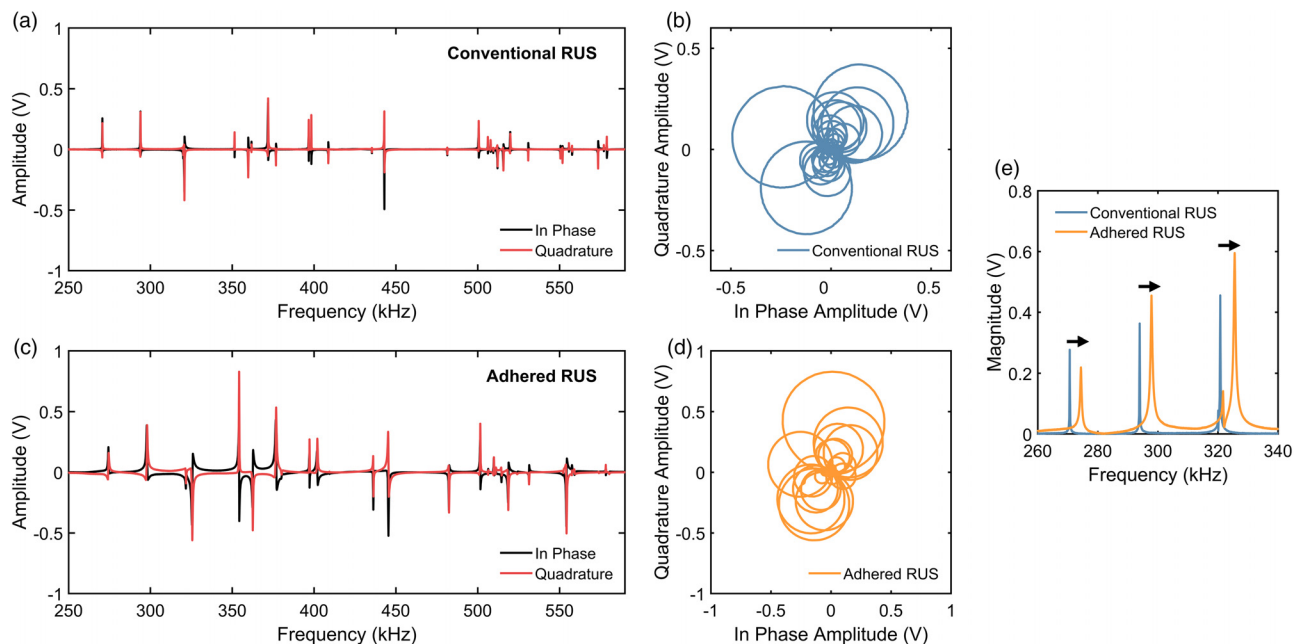


FIG. 3. (Color online) Resonant ultrasound spectra acquired on a high-density tungsten alloy sample (a),(b) with conventional RUS in which the sample is lightly held between the two transducers and (c),(d) with the sample and transducers coupled with a small amount of adhesive. (e) The primary effects are a shift in the resonant frequencies, increase in width, and increase in amplitude. Note the excitation voltage is factor of 3 smaller when the adhesive is used.

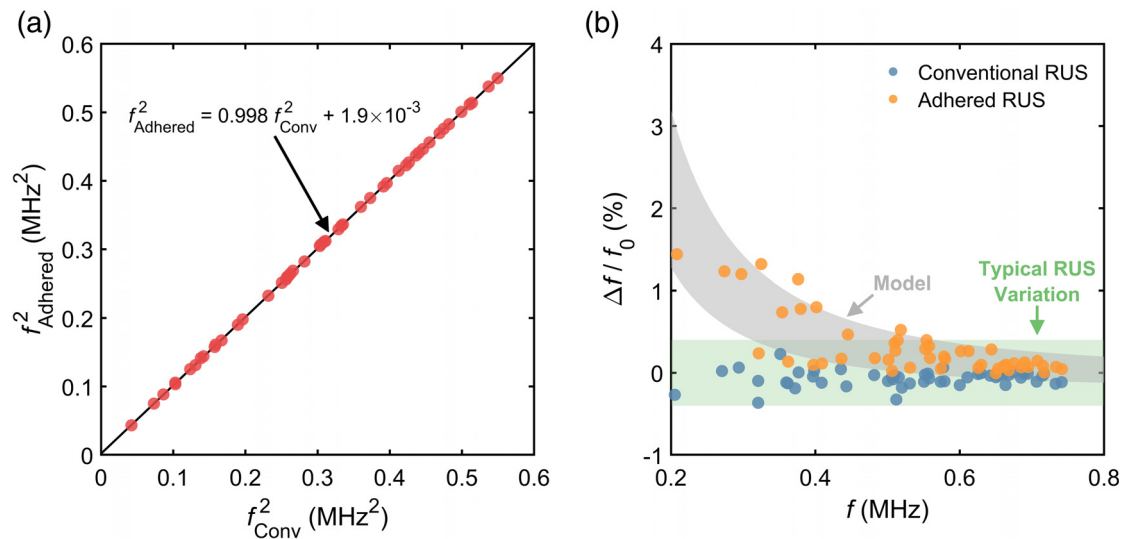


FIG. 4. (Color online) Changes in the resonant frequencies from the adhesive. (a) The square of the resonant frequencies measured with the adhesive (f_{Adhered}^2) scales linearly with the square of the resonant frequencies measured without the adhesive (f_{Conv}^2), as predicted by Eq. (1). (b) The change in the resonant frequencies with the adhesive relative to the resonant frequencies without the adhesive (orange) is larger than, but comparable to, the typical variation in conventional RUS from varying the placement of the sample between the transducers (blue). The changes resulting from the adhesive are captured by our model [gray shaded region corresponds to 95% confidence interval of the fit in (a)] and fall within typical RUS variations at high frequencies (green shaded region).

of elastic constants determined from conventional RUS after adhesive effects have been accounted for. Table I and Fig. 5 provide comparisons of the isotropic elastic constants obtained from measurements on a high-density tungsten alloy sample with and without an adhesive. Elastic constants were determined via Visscher's method²³ with the same number of resonances used for the inversions. Measured frequencies and fit results are provided in Tables II–IV in Appendix B.

We first note that for the un-adhered case, the elastic constants are well characterized with either 20 or 52 resonances. However, as shown in Fig. 5 and Table I, when elastic constants are determined from the first 20 as-measured frequencies with an adhered sample, the elastic constants differ substantially from the elastic constants obtained with conventional RUS. The situation is improved by increasing the number of frequencies included in the inversion to the first 52 as-measured frequencies, but the resulting elastic constants are still clearly outside the uncertainty of the elastic constants found with conventional RUS. This improvement in accuracy from increasing the number of resonant frequencies included in the inversion arises because the

adhesive-induced shift in the resonant frequencies becomes relatively smaller at higher frequencies (Fig. 4).

In contrast, when the frequencies measured with the adhesive are corrected using the fit parameters found from applying Eq. (1), quantitative agreement is obtained with the elastic constants determined by conventional RUS (Fig. 5). More specifically, the correction process requires finding the fit parameters A and B according to $f'^2 = Af_0^2 + B$, where f' are the as-measured resonant frequencies with an adhesive applied to the sample and f_0 are the free resonant frequencies of the sample measured with conventional RUS. The agreement between the elastic constants found with this method and those determined from conventional RUS holds when either the first 20 or 52 resonant frequencies are included in the inversion. Note, the small differences in c_{11} between inversions using the first 20 and 52 frequencies is the result of better c_{11} representation in the larger number of frequencies, whereas c_{44} has sufficient representation in every resonance (see Appendix B).

Having demonstrated elastic constants can be determined to within the uncertainty of conventional RUS when

TABLE I. Room temperature elastic constants determined from the first 20 and 52 resonances on a high-density tungsten alloy rectangular parallelepiped sample assuming an isotropic fit (2 degrees of freedom). Conventional RUS refers to inversions on resonance spectra obtained without an adhesive. Adhered RUS (As Measured) corresponds to elastic constants computed from the as-measured resonant frequencies obtained with an adhesive. Adhered RUS (Corrected) provides elastic constants which have been corrected for adhesive effects by shifting frequencies according to the Eq. (1) fit shown in Fig. 4(a).

	Conventional RUS		Adhered RUS (As Measured)		Adhered RUS (Corrected)	
Number of resonant frequencies	20	52	20	52	20	52
c_{11} (GPa)	442.7 ± 3.3	447.2 ± 2.0	424.7 ± 7.2	440.3 ± 4.5	441.3 ± 4.9	445.6 ± 3.3
c_{44} (GPa)	134.7 ± 0.1	134.4 ± 0.1	137.1 ± 0.3	135.7 ± 0.2	134.6 ± 0.2	134.5 ± 0.1
B (GPa)	263.1 ± 3.3	268.0 ± 2.0	241.9 ± 7.2	259.3 ± 4.5	261.9 ± 4.9	266.3 ± 3.3
ν	0.393 ± 0.002	0.399 ± 0.001	0.364 ± 0.006	0.387 ± 0.003	0.392 ± 0.003	0.397 ± 0.002

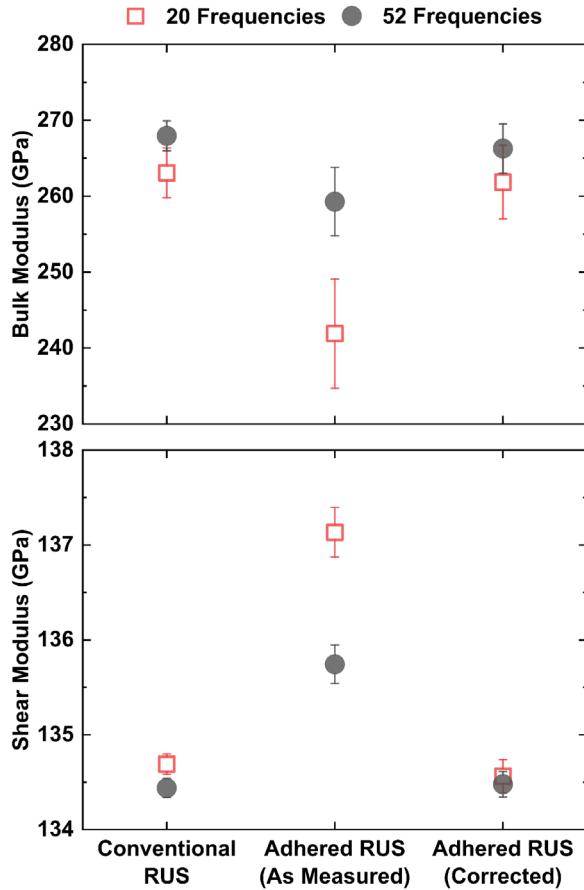


FIG. 5. (Color online) Comparison of the bulk and shear moduli of a tungsten alloy rectangular parallelepiped sample determined with conventional RUS, the as-measured frequencies obtained when an adhesive is used, and the frequencies measured with the adhesive which have been corrected according to Eq. (1). In all cases, inversions were performed with the first 52 (gray closed circles) and 20 (red open squares) resonant frequencies. Elastic constants determined from the as-measured resonant frequencies obtained when an adhesive is used are outside the uncertainty of elastic constants determined by conventional RUS. Correcting the frequencies measured with the adhesive according to Eq. (1) yields quantitative agreement with conventional RUS. Values are provided in Table I.

an adhesive is used, we now turn to using our modified RUS approach for temperature- and magnetic field-dependent studies. The application of RUS in these contexts is highly desirable because elastic constants in the vicinity of phase transitions are renormalized by strain-order parameter and electron-phonon couplings.^{6–8} Such deviations from typical elastic constant behavior⁴⁸ can be used to understand the nature of phase transitions and place constraints on order parameter symmetries.^{6,16,49} As previously discussed, the principal effect of adhering a sample to a transducer is a constant shift in the resonant frequencies to higher frequencies [Eq. (1)]. Consequently, changes in resonant frequencies (and elastic constants), which are the important quantity in temperature- and magnetic field-dependent studies,⁴⁶ should be largely unaffected by the adhesive. We experimentally confirm this is the case using a single crystal of SrTiO₃ (Appendix C) and polycrystalline Gd. Gd was selected to test our approach because it has well-known

magnetic phase transitions as a function of temperature and applied magnetic field, including a paramagnetic-ferromagnetic transition and a spin-reorientation transition at zero applied magnetic field (e.g., see Ref. 41, and references therein). These phase transitions are an ideal test because they involve large changes in magnetization, causing the sample to experience substantial torque⁵⁰ which tends to knock samples out of transducers in conventional RUS setups.³⁵ Indeed, every attempt to apply a magnetic field to Gd samples in conventional RUS without an adhesive led to the sample flying away at low and irreproducible fields ($\mu_0 H \sim 0.1$ T).

Figure 6 shows the temperature-dependent behavior of a resonant frequency of a polycrystalline Gd cylinder at fixed magnetic fields under free resonance conditions and with an adhesive used to affix the sample to the transducers. Focusing first on the zero applied magnetic field data, these measurements demonstrate that both the ferromagnetic and spin-reorientation transitions are clearly resolved in the adhered sample with transition temperatures that agree with the literature.⁴¹ Furthermore, the temperature dependence at zero applied magnetic field is essentially unchanged by the adhesive: maxima and minima occur at the same temperatures and the data show similar concavity [Fig. 6(b)]. The acoustic quality factors, while reduced, are still sufficient to observe changes in ultrasonic attenuation at phase transitions. See Appendix D for additional discussion of the effects of the adhesive on ultrasonic attenuation. Note, the good agreement between the temperature dependences measured with and without adhesive in Fig. 6(b) indicates the adhesive is not substantially changing the relative amounts of different elastic constants in each normal mode and provides additional evidence that the dominant effect of the adhesive in these measurements is a rigid shift in the resonant frequencies. There also do not appear to be any thermal-strain effects associated with the adhesive which alter the temperature dependence of the sample's resonant frequency spectra, likely because the amount of adhesive is minimal. That being said, in general it is important to characterize the temperature-dependent properties of the adhesive for a particular sample/application over the measurement conditions. By maintaining the assumptions which led to Eq. (1), i.e., $f^2 - f_0^2 \approx 0$, contributions to the temperature-dependent elastic response from the adhesive should be minimized (Fig. 6). Additionally, the adhesive may improve thermal conductivity to the sample which can benefit very low temperature RUS applications which commonly face sample thermalization difficulties.⁵¹

Measuring the temperature-dependent behavior of a resonant frequency at a fixed applied magnetic field of 2 T on the adhered samples shows both the ferromagnetic and spin-reorientation phase transitions are suppressed, in agreement with the literature.⁴¹ Comparable measurements with conventional RUS at 2 T could not be performed because the sample flew away at substantially smaller applied magnetic fields (<0.1 T). To further demonstrate our adhesive-enabled RUS capabilities at high magnetic fields, we

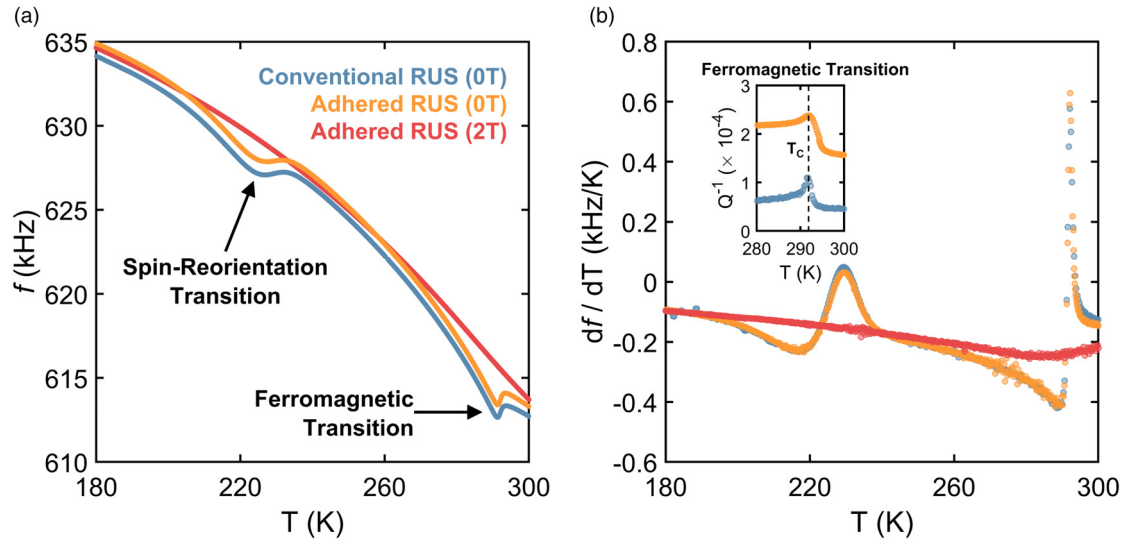


FIG. 6. (Color online) Temperature-dependence of a resonant frequency of a polycrystalline Gd cylinder at different applied magnetic fields with and without the sample adhered to the transducers. (a) The magnetic phase transitions in Gd at zero applied magnetic field are clearly resolved as relative minima with (orange) and without (blue) adhesive. These phase transitions are suppressed by the application of a 2 T magnetic field (red). (b) The derivative of the resonant frequency with respect to temperature emphasizes that changes in the resonant frequency in the vicinity of both phase transitions are only minimally affected by the adhesive. Evidence for the phase transitions is also present in ultrasonic attenuation, even with the adhesive (ferromagnetic transition, inset).

measured the same resonant frequency shown in Fig. 6 as a function of magnetic field up to 14 T at a fixed temperature of 260 K (Fig. 7). This temperature places the sample below the ferromagnetic transition and above the spin-reorientation transition, a region of phase space where it is known that Gd undergoes a phase transition at $\mu_0 H \approx 0.4$ T.⁴¹ As shown in Fig. 7, we can perform high signal-to-noise RUS measurements up to 14 T and identify signatures of this phase transition⁴¹ in both resonant frequencies and ultrasonic attenuation. Therefore, this new RUS approach provides the same sensitivity to phase

transitions at zero field compared to conventional RUS and enables reproducible RUS measurements in large magnetic fields.

IV. DISCUSSION

RUS is an ideal technique to study elasticity and determine phase diagrams under extreme environments because of its high precision and accuracy, and its ability to simultaneously determine the entire elastic constant tensor from a single measurement. The latter benefit is especially significant because

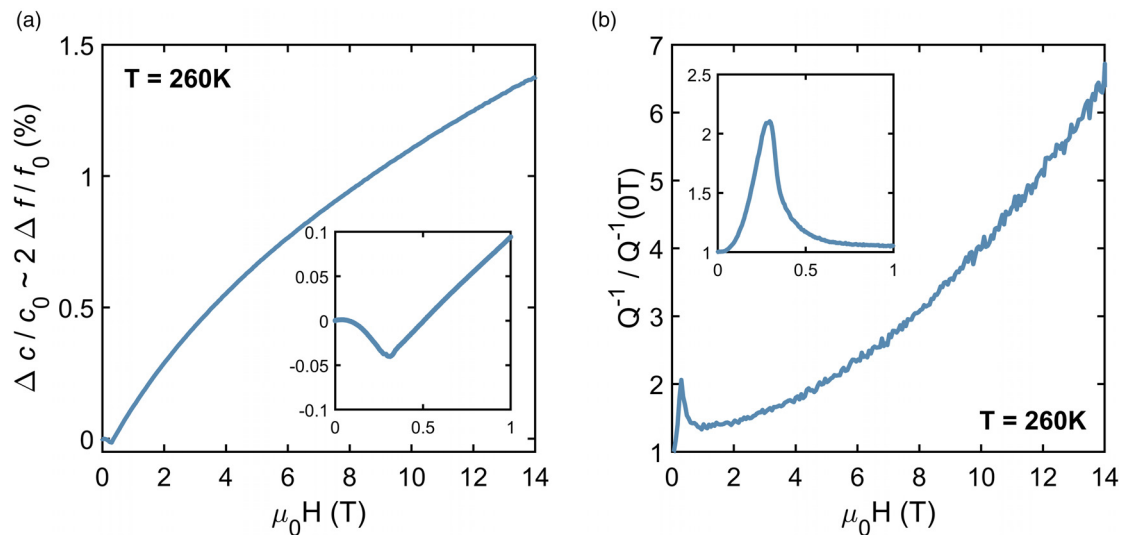


FIG. 7. (Color online) Magnetic field dependence of a resonant frequency of a polycrystalline Gd cylinder measured with the sample adhered to the transducers up to 14 T at fixed temperature. (a) The relative change in the frequency (proportional to the relative change in the elastic constants) shows high signal-to-noise up to 14 T as well as evidence for a field-induced phase transition at low fields. (b) The change in the ultrasonic attenuation up to 14 T also has high signal-to-noise, enabling the measurement of a clear maximum at the low-field phase transition. Insets are higher resolution scans acquired with a slower magnetic field ramp rate showing the behavior in the vicinity of the field-induced phase transition.

obtaining comparable datasets with alternative approaches (e.g., pulse-echo ultrasound techniques⁶) requires combining multiple samples and measurements, introducing the possibility of large systematic uncertainties. We have shown that the primary challenge in utilizing RUS in extreme environments, namely, sample stability, is readily addressed by adhering a sample to the transducers in an otherwise conventional RUS experiment. The adhesive improves sample stability and orientation control, allowing for reproducible RUS in extreme environments such as high magnetic fields. The presence of the adhesive does not reduce sensitivity to phase transitions, as demonstrated by the fact that phase transitions are readily observed in both elastic constants and ultrasonic attenuation. Modelling the adhesive effects even allows one to obtain elastic constants in excellent quantitative agreement with conventional RUS. Furthermore, we anticipate our findings may also be beneficial to other RUS approaches such as non-contact methods.³⁰

Beyond the opportunities to perform RUS measurements in new environments and regions of phase space, the use of the adhesive may improve RUS investigations under historically challenging conditions and on difficult samples. As shown in Figs. 2 and 3, the excitation voltage used in RUS experiments

when an adhesive is present must be substantially reduced to avoid saturating detection electronics. This enhanced sample-transducer coupling can assist in the study of polymeric and geological materials which are often weak resonators^{43,44} while maintaining pseudo-free resonance conditions. The ability to tune sample-transducer coupling may also find applications in high-pressure RUS experiments, where reduced resonant frequency amplitudes and increased ultrasonic attenuation owing to the pressure medium are major obstacles.⁵² Last, the adhesive acts as a thermal link to the sample which can improve thermalization and facilitate ultralow temperature RUS measurements.⁵¹

This approach also provides an opportunity to investigate the properties of the adhesive materials themselves (e.g., to understand the efficacy of transducer couplants⁵³). As shown in Fig. 8, changes in the resonant frequencies of a sample as a function of time reflect the dynamics of the curing process. The adhesive stiffens as it cures, shifting the resonant frequencies of the sample to higher values, increasing the resonant quality factor, and enhancing the coupling to the transducers. By pairing time-dependent RUS measurements on adhered samples with controlled external

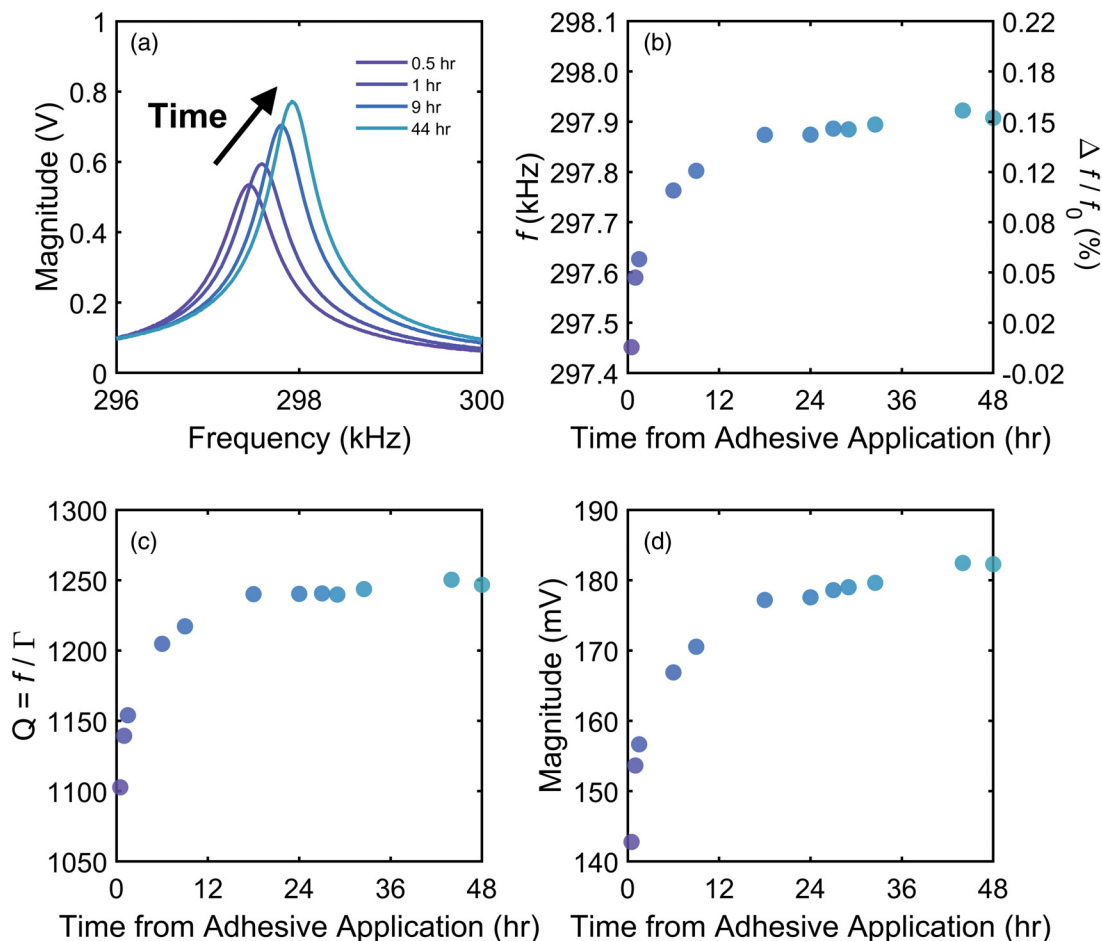


FIG. 8. (Color online) Adhesive curing effects measured by resonant ultrasound spectroscopy. (a) Changes in the resonant response of a high-density tungsten alloy sample from the time of the adhesive application. The (b) resonant frequency, (c) quality factor (frequency / width), and (d) magnitude of the resonance all increase with time, saturating once the adhesive has cured.

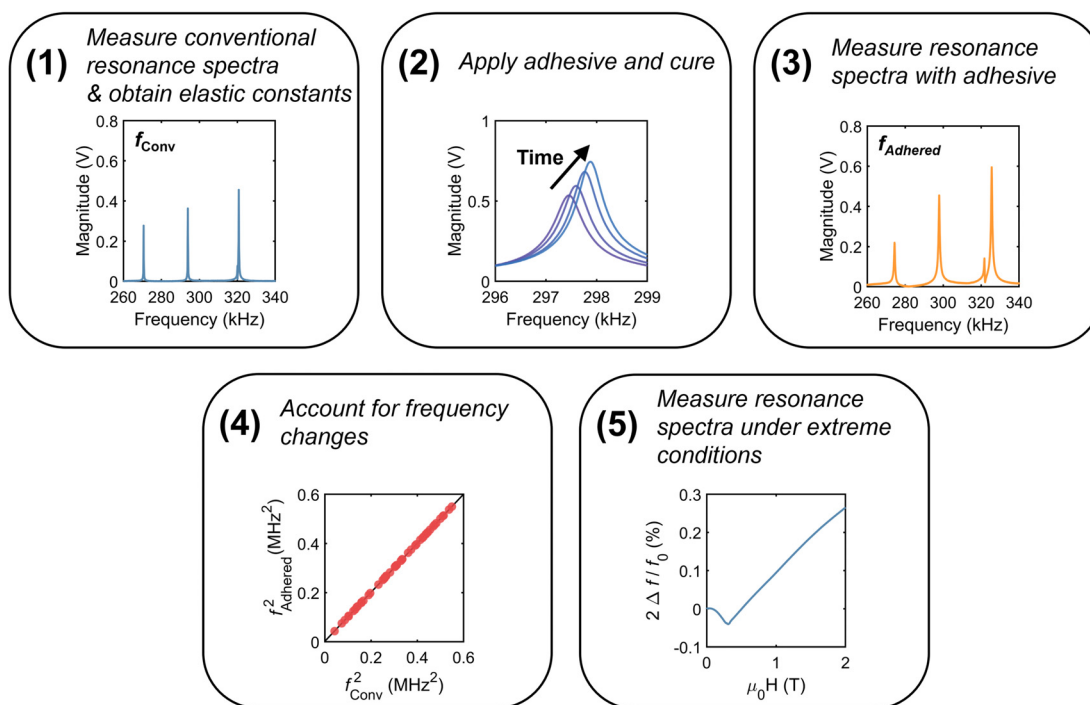


FIG. 9. (Color online) Summary of a typical workflow for performing RUS measurements in high magnetic fields with the sample adhered to the transducers.

conditions (gas composition, humidity levels, light exposure, etc.), intrinsic and dynamic polymeric properties can be studied. For the purposes of these measurements, the data in Fig. 8 also demonstrate the importance of waiting for the adhesive to completely cure before beginning measurements. If the adhesive is not fully cured, changes in the adhesive may be erroneously attributed to the sample.

Finally, we provide a typical workflow for performing RUS measurements in high magnetic fields (or other extreme environments) where the sample has been adhered to the transducers (Fig. 9). Naturally, details are sample and adhesive dependent, but we have found the following steps are a useful guideline.

- (1) Measure a resonant frequency spectrum with conventional RUS at ambient conditions. Ensure a high-quality inversion can be obtained with results which are not especially sensitive to transducer placement.
- (2) Apply a sample-compatible adhesive and measure changes in the resonance spectrum. Wait until there are no significant changes in the resonance spectrum. Ideally, measure the changes as a function of time to check the adhesive has fully cured, at least the first time a particular adhesive is used.
- (3) Once the adhesive has cured, measure a resonance spectrum with the adhesive at the same conditions as (1).
- (4) Check that frequency shifts before and after the application of the adhesive are small ($\sim 1\%$) and correct the resonant frequencies measured with the adhesive according to the Eq. (1). Confirm inversion with corrected frequencies gives reasonable elastic constants. If not, re-adhere sample.

- (5) Perform RUS measurements. Analyze relative changes in elastic constants from relative changes in frequency⁴⁶ or changes in absolute values with corrected frequencies. Remove adhesive if desired.

V. CONCLUSION

In this work, we demonstrated how to perform quantitative elastic constant measurements and phase boundary determination under extreme conditions, such as high magnetic fields, using a variation of resonant ultrasound spectroscopy in which the sample is adhered to ultrasonic transducers. We outlined the experimental conditions in which the adhesive can be treated as a perturbation to the free resonance condition and show that, in this regime, the adhesive acts to shift the resonant frequencies by a constant value. These effects can be accounted for by a simple model such that elastic constants can be efficiently obtained with the same accuracy and sensitivity to phase transitions as conventional RUS, but with substantial improvements in sample stability and controlled sample orientation because of the adhesive. This development facilitates the application of RUS to new, extreme environments including reproducible measurements of elastic constants and ultrasonic attenuation in magnetic fields up to 14 T.

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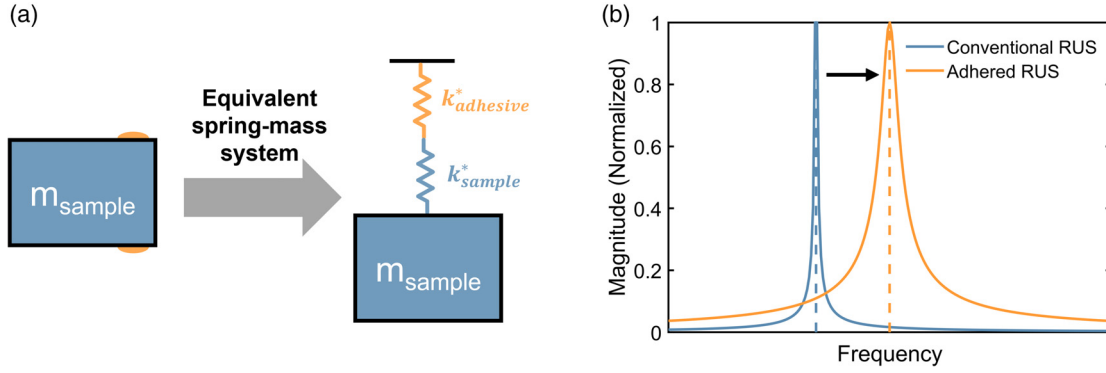


FIG. 10. (Color online) (a) Depiction of effective spring-mass system describing a sample coupled to transducers with an adhesive. (b) Simulated effects of the adhesive on the resonant frequency and resonance width.

Florida). We thank Tannor T. J. Munroe and Fedor Balakirev for assistance with sample preparation and instrumentation.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts of interest to disclose.

DATA AVAILABILITY

Data used in this work are provided in [Appendix B](#). Additional data available upon request.

APPENDIX A: EFFECTIVE SPRING-MASS MODEL FOR ADHESIVE EFFECTS

To model the effects of the adhesive on the resonant behavior of a sample, we consider the sample to be a one-dimensional driven harmonic oscillator^{9,12} of mass m_s whose displacement x under an applied force F_a is governed by the differential equation

$$m_s \ddot{x} = -k^* x + F_a. \quad (\text{A1})$$

We assume the sample and adhesive act as two complex springs in series with one spring representing the anelastic response of the sample (k_s^*) and the other representing the anelastic response of the adhesive (k_g^*) (see Fig. 10). Then, the effective spring constant of this system is

$$k^* = k_s^* + k_g^* = k_s + k_g + i(k_s \tan \phi_s + k_g \tan \phi_g), \quad (\text{A2})$$

where ϕ_s and ϕ_g are the loss angles of the sample and adhesive, respectively.⁹

If we take the applied force F_a to be a sinusoidal wave with amplitude F_0 , frequency ω , and phase angle θ ,

$$F_a = F_0 e^{i(\omega t + \theta)} \quad (\text{A3})$$

and assume the displacements x to be plane waves with amplitude x_0 and frequency ω ,

$$x = x_0 e^{i\omega t}, \quad (\text{A4})$$

the steady-state amplitude response of the system is given by

$$|x_0|^2 = \frac{\left(\frac{F_0}{m_s}\right)^2}{(\omega^2 - \omega'^2)^2 + \left(\omega_0^2 \tan \phi_s + (\omega^2 - \omega_0^2) \tan \phi_g\right)^2}, \quad (\text{A5})$$

where

$$\omega'^2 = \omega_0^2 + \frac{k_g}{m_s} \quad (\text{A6})$$

is resonant frequency of the sample-adhesive system and

$$\omega_0^2 = \frac{k_s}{m_s} \quad (\text{A7})$$

is the resonant frequency of the sample by itself. Therefore, the adhesive effectively acts like an additional force acting on the sample which enters Eq. (A5) through the resonant frequency of the sample-adhesive system, ω'^2 , shown in Eq. (A6).²⁹ Note, the model does not penalize the decrease in Q caused by having larger contact area. To recover the form of Eq. (1) from Eq. (A6) we use $\omega = 2\pi f$ and the following relationship between a spring constant (k), Young's modulus (Y), cross-sectional area (A), thickness (t), volume (V), mass (m), and density (ρ):

$$k = Y \frac{A}{t} = Y \frac{V}{t^2} = Y \frac{m}{\rho t^2}. \quad (\text{A8})$$

Note, it may be more appropriate to use another elastic modulus instead of Young's modulus in Eq. (A8) depending on the details of the adhesive shape and the eigenmodes of the sample.

APPENDIX B: ROOM TEMPERATURE ELASTIC CONSTANT DETERMINATION OF TUNGSTEN ALLOY SAMPLE

The tungsten alloy sample, shaped into a rectangular parallelepiped with dimensions of 4.024(5) mm $\times$ 5.036(5) mm $\times$ 6.038(5) mm, was used for quantitative analyses. Its mass

TABLE II. Example output from RUS inversion code (Ref. 13) of a typical fit to the resonant frequencies of a high density tungsten alloy sample using conventional RUS (i.e., no adhesive). The RMS error of the fit to the experimental frequencies was 0.184%. Elastic moduli are given in Table I.

Resonance number	Experimental frequency (MHz)	Calculated frequency (MHz)	Error (%)	Fit weight	k	i	$\ln(f)/\ln(c_{11})$	$\ln(f)/\ln(c_{44})$
1	0.205 295	0.204 164	−0.55	1	4	1	0.000 6	0.999 4
2	0.270 658	0.270 049	−0.22	1	1	2	0.123 94	0.876 06
3	0.293 93	0.293 102	−0.28	1	7	2	0.156 94	0.843 06
4	0.320 761	0.319 909	−0.27	1	4	2	0.002 2	0.997 8
5	0.320 761	0.320 773	0	1	8	2	0.018 8	0.981 2
6	0.351 415	0.352 137	0.21	1	5	1	0.043 95	0.956 05
7	0	0.359 998	0	0	2	2	0.018 1	0.981 9
8	0.361 775	0.361 756	−0.01	1	1	3	0.308 33	0.691 67
9	0.371 888	0.370 562	−0.36	1	3	2	0.068 79	0.931 21
10	0.376 741	0.377 05	0.08	1	6	2	0.111 62	0.888 38
11	0.396 743	0.397 787	0.26	1	3	3	0.059 51	0.940 49
12	0.398 319	0.398 178	−0.04	1	5	2	0.084 75	0.915 25
13	0.408 677	0.408 799	0.03	1	2	3	0.157 15	0.842 85
14	0.435 367	0.436 766	0.32	1	5	3	0.083 03	0.916 97
15	0.442 965	0.442 493	−0.11	1	3	4	0.041 18	0.958 82
16	0.481 437	0.481 822	0.08	1	7	3	0.113 94	0.886 06
17	0	0.500 894	0	0	6	3	0.064 03	0.935 97
18	0.500 596	0.501 015	0.08	1	4	3	0.077 65	0.922 35
19	0.506 186	0.506 59	0.08	1	8	3	0.130 25	0.869 75
20	0.507 854	0.508 231	0.07	1	6	4	0.191 36	0.808 64
21	0.509 48	0.508 773	−0.14	1	5	4	0.342 64	0.657 36
22	0.512 125	0.510 338	−0.35	1	1	4	0.117 13	0.882 87
23	0.515 661	0.516 021	0.07	1	7	4	0.152 12	0.847 88
24	0	0.519 504	0	0	2	4	0.038 4	0.961 6
25	0.530 859	0.530 605	−0.05	1	5	5	0.325 67	0.674 33
26	0.550 516	0.550 368	−0.03	1	7	5	0.180 63	0.819 37
27	0.551 794	0.552 967	0.21	1	1	5	0.069 79	0.930 21
28	0.555 715	0.555 561	−0.03	1	8	4	0.052 09	0.947 91
29	0.557 641	0.557 917	0.05	1	6	5	0.186 22	0.813 78
30	0.573 558	0.573 751	0.03	1	1	6	0.067 97	0.932 03
31	0.577 055	0.577 862	0.14	1	2	5	0.109 34	0.890 66
32	0.578 901	0.578 424	−0.08	1	6	6	0.110 17	0.889 83
33	0.600 009	0.598 429	−0.26	1	8	5	0.144 52	0.855 48
34	0.610 662	0.610 476	−0.03	1	4	4	0.012 12	0.987 88
35	0.625 488	0.627 404	0.31	1	1	7	0.106 67	0.893 33
36	0.629 141	0.630 81	0.27	1	7	6	0.081 14	0.918 86
37	0.642 074	0.643 295	0.19	1	3	5	0.066 35	0.933 65
38	0.650 111	0.650 65	0.08	1	5	6	0.194 75	0.805 25
39	0.653 063	0.652 997	−0.01	1	5	7	0.615 93	0.384 07
40	0.660 408	0.661 799	0.21	1	2	6	0.125 04	0.874 96
41	0.663 406	0.662 44	−0.15	1	4	5	0.117 33	0.882 67
42	0.667 864	0.668 06	0.03	1	8	6	0.131 22	0.868 78
43	0.674 55	0.675 296	0.11	1	7	7	0.081 22	0.918 78
44	0.684 793	0.685 104	0.05	1	6	7	0.109 43	0.890 57
45	0.689	0.690 321	0.19	1	3	6	0.114 52	0.885 48
46	0.694 245	0.695 108	0.12	1	5	8	0.252 8	0.747 2
47	0.706 566	0.704 873	−0.24	1	7	8	0.149 64	0.850 36
48	0.714 611	0.715 059	0.06	1	4	6	0.079 04	0.920 96
49	0.716 844	0.717 2	0.05	1	2	7	0.052 77	0.947 23
50	0	0.718 234	0	0	8	7	0.055 06	0.944 94
51	0.732 772	0.732 389	−0.05	1	6	8	0.080 61	0.919 39
52	0.741 196	0.739 907	−0.17	1	5	9	0.111 44	0.888 56

TABLE III. Example output from RUS inversion code (Ref. 13) of a typical fit to the resonant frequencies of a high density tungsten alloy sample which has been adhered to the transducers. Frequencies have been corrected according to Eq. (1). The RMS error of the fit to the experimental frequencies was 0.294%. Elastic moduli are given in Table I.

Resonance number	Experimental frequency (MHz)	Calculated frequency (MHz)	Error (%)	Fit weight	k	i	$\text{dln}(f)/\text{dln}(c_{11})$	$\text{dln}(f)/\text{dln}(c_{44})$
1	0.203 87	0.204 194	0.16	1	4	1	0.000 61	0.999 39
2	0.270 805	0.270 022	−0.29	1	1	2	0.125 25	0.874 75
3	0.294 569	0.293 054	−0.51	1	7	2	0.158 48	0.841 52
4	0.318 908	0.319 955	0.33	1	4	2	0.002 22	0.997 78
5	0.322 416	0.320 808	−0.5	1	8	2	0.018 99	0.981 01
6	0.351 698	0.352 158	0.13	1	5	1	0.044 57	0.955 43
7	0	0.360 038	0	0	2	2	0.018 29	0.981 71
8	0.360 035	0.361 588	0.43	1	1	3	0.310 7	0.689 3
9	0.373 999	0.370 566	−0.92	1	3	2	0.069 6	0.930 4
10	0.377 578	0.377 021	−0.15	1	6	2	0.112 9	0.887 1
11	0.395 165	0.397 798	0.67	1	3	3	0.059 94	0.940 06
12	0.399 568	0.398 169	−0.35	1	5	2	0.085 84	0.914 16
13	0.407 262	0.408 732	0.36	1	2	3	0.158 65	0.841 35
14	0.434 409	0.436 758	0.54	1	5	3	0.084 26	0.915 74
15	0.443 38	0.442 522	−0.19	1	3	4	0.041 42	0.958 58
16	0.480 854	0.481 784	0.19	1	7	3	0.115 67	0.884 33
17	0	0.500 902	0	0	6	3	0.066 76	0.933 24
18	0.500 042	0.501 011	0.19	1	4	3	0.078 46	0.921 54
19	0.504 976	0.506 534	0.31	1	8	3	0.131 77	0.868 23
20	0.508 384	0.508 114	−0.05	1	6	4	0.191 11	0.808 89
21	0.509 546	0.508 502	−0.2	1	5	4	0.345 1	0.654 9
22	0.512 874	0.510 294	−0.5	1	1	4	0.118 19	0.881 81
23	0.517 086	0.515 941	−0.22	1	7	4	0.152 93	0.847 07
24	0	0.519 541	0	0	2	4	0.038 7	0.961 3
25	0.529 983	0.530 34	0.07	1	5	5	0.328 66	0.671 34
26	0.551 008	0.550 251	−0.14	1	7	5	0.182 15	0.817 85
27	0.552 878	0.552 971	0.02	1	1	5	0.070 38	0.929 62
28	0.556 423	0.555 584	−0.15	1	8	4	0.052 58	0.947 42
29	0.557 555	0.557 793	0.04	1	6	5	0.188 53	0.811 47
30	0.572 822	0.573 758	0.16	1	1	6	0.068 38	0.931 62
31	0.577 189	0.577 822	0.11	1	2	5	0.110 36	0.889 64
32	0.578 865	0.578 383	−0.08	1	6	6	0.110 61	0.889 39
33	0.600 667	0.598 345	−0.39	1	8	5	0.145 8	0.854 2
34	0.611 391	0.610 551	−0.14	1	4	4	0.012 26	0.987 74
35	0.625 033	0.627 363	0.37	1	1	7	0.107 4	0.892 6
36	0.628 932	0.630 8	0.3	1	7	6	0.081 97	0.918 03
37	0.643 131	0.643 304	0.03	1	3	5	0.067 03	0.932 97
38	0.649 327	0.650 467	0.18	1	5	6	0.244 81	0.755 19
39	0.652 665	0.652 32	−0.05	1	5	7	0.577 65	0.422 35
40	0.660 225	0.661 731	0.23	1	2	6	0.126 19	0.873 81
41	0.663 354	0.662 382	−0.15	1	4	5	0.118 46	0.881 54
42	0.667 38	0.667 984	0.09	1	8	6	0.132 27	0.867 73
43	0.674 665	0.675 286	0.09	1	7	7	0.081 78	0.918 22
44	0.684 624	0.685 056	0.06	1	6	7	0.110 3	0.889 7
45	0.689 252	0.690 265	0.15	1	3	6	0.115 85	0.884 15
46	0.694 13	0.694 863	0.11	1	5	8	0.251 59	0.748 41
47	0.707 025	0.704 767	−0.32	1	7	8	0.150 98	0.849 02
48	0.714 673	0.715 051	0.05	1	4	6	0.079 89	0.920 11
49	0.716 337	0.717 229	0.12	1	2	7	0.053 17	0.946 83
50	0	0.718 261	0	0	8	7	0.055 91	0.944 09
51	0.732 805	0.732 379	−0.06	1	6	8	0.081 31	0.918 69
52	0.741 066	0.739 852	−0.16	1	5	9	0.112 13	0.887 87

TABLE IV. Example output from RUS inversion code (Ref. 13) of a typical fit to the resonant frequencies of a high density tungsten alloy sample which has been adhered to the transducers. Frequencies are un-corrected for adhesive effects. The RMS error of the fit to the experimental frequencies was 0.472%. Elastic moduli are given in Table I.

Resonance number	Experimental frequency (MHz)	Calculated frequency (MHz)	Error (%)	Fit weight	k	i	$\text{dln}(f)/\text{dln}(c_{11})$	$\text{dln}(f)/\text{dln}(c_{44})$
1	0.208 25	0.205 15	-1.49	1	4	1	0.000 63	0.999 37
2	0.273 99	0.270 915	-1.12	1	1	2	0.132 69	0.867 31
3	0.297 451	0.293 917	-1.19	1	7	2	0.167 18	0.832 82
4	0.321 521	0.321 447	-0.02	1	4	2	0.002 33	0.997 67
5	0.324 993	0.322 245	-0.85	1	8	2	0.020 07	0.979 93
6	0.354 001	0.353 634	-0.1	1	5	1	0.048 17	0.951 83
7	0	0.361 653	0	0	2	2	0.019 37	0.980 63
8	0.362 268	0.362 056	-0.06	1	1	3	0.323 93	0.676 07
9	0.376 118	0.372 018	-1.09	1	3	2	0.074 24	0.925 76
10	0.379 67	0.378 319	-0.36	1	6	2	0.120 18	0.879 82
11	0.397 126	0.399 403	0.57	1	3	3	0.062 29	0.937 71
12	0.401 499	0.399 657	-0.46	1	5	2	0.092 12	0.907 88
13	0.409 14	0.409 935	0.19	1	2	3	0.167 1	0.832 9
14	0.436 113	0.438 395	0.52	1	5	3	0.091 38	0.908 62
15	0.445 03	0.444 397	-0.14	1	3	4	0.042 75	0.957 25
16	0.482 297	0.483 421	0.23	1	7	3	0.125 64	0.874 36
17	0	0.502 847	0	0	6	3	0.084 95	0.915 05
18	0.501 389	0.502 926	0.31	1	4	3	0.083 06	0.916 94
19	0.506 299	0.508 171	0.37	1	8	3	0.140 4	0.859 6
20	0.509 691	0.508 973	-0.14	1	5	4	0.358 74	0.641 26
21	0.510 847	0.509 466	-0.27	1	6	4	0.186 91	0.813 09
22	0.514 16	0.512 025	-0.42	1	1	4	0.124 15	0.875 85
23	0.518 352	0.517 504	-0.16	1	7	4	0.157 3	0.842 7
24	0	0.521 757	0	0	2	4	0.040 37	0.959 63
25	0.531 191	0.530 915	-0.05	1	5	5	0.345 39	0.654 61
26	0.552 125	0.551 733	-0.07	1	7	5	0.190 67	0.809 33
27	0.553 987	0.555 138	0.21	1	1	5	0.073 7	0.926 3
28	0.557 517	0.557 868	0.06	1	8	4	0.055 39	0.944 61
29	0.558 644	0.559 244	0.11	1	6	5	0.201 48	0.798 52
30	0.573 849	0.576 021	0.38	1	1	6	0.070 66	0.929 34
31	0.578 199	0.579 83	0.28	1	2	5	0.116 11	0.883 89
32	0.579 868	0.580 402	0.09	1	6	6	0.113 18	0.886 82
33	0.601 587	0.600 194	-0.23	1	8	5	0.153 01	0.846 99
34	0.612 272	0.613 33	0.17	1	4	4	0.013 08	0.986 92
35	0.625 866	0.629 569	0.59	1	1	7	0.111 5	0.888 5
36	0.629 751	0.633 189	0.55	1	7	6	0.086 63	0.913 37
37	0.643 901	0.645 845	0.3	1	3	5	0.070 93	0.929 07
38	0.650 076	0.649 842	-0.04	1	5	6	0.713 01	0.286 99
39	0.653 403	0.653 108	-0.05	1	5	7	0.172 7	0.827 3
40	0.660 938	0.663 917	0.45	1	2	6	0.132 67	0.867 33
41	0.664 057	0.664 626	0.09	1	4	5	0.124 84	0.875 16
42	0.668 07	0.670 149	0.31	1	8	6	0.138 15	0.861 85
43	0.675 331	0.677 849	0.37	1	7	7	0.084 9	0.915 1
44	0.685 26	0.687 441	0.32	1	6	7	0.115 14	0.884 86
45	0.689 873	0.692 618	0.4	1	3	6	0.123 36	0.876 64
46	0.694 736	0.696 275	0.22	1	5	8	0.245 15	0.754 85
47	0.707 592	0.706 905	-0.1	1	7	8	0.158 48	0.841 52
48	0.715 218	0.717 773	0.36	1	4	6	0.084 73	0.915 27
49	0.716 876	0.720 175	0.46	1	2	7	0.055 42	0.944 58
50	0	0.721 18	0	0	8	7	0.060 84	0.939 16
51	0.733 297	0.735 16	0.25	1	6	8	0.085 22	0.914 78
52	0.741 534	0.742 418	0.12	1	5	9	0.115 99	0.884 01

TABLE V. Room temperature elastic constants determined from the first 33 resonances on a SrTiO_3 single crystal cylinder assuming a cubic fit (3 degrees of freedom). Conventional RUS refers to inversions on resonance spectra obtained without an adhesive. Adhered RUS (As Measured) corresponds to elastic constants computed from the as-measured resonant frequencies obtained with an adhesive. Adhered RUS (Corrected) provides elastic constants which have been corrected for adhesive effects by shifting frequencies according to Eq. (1).

	Conventional RUS	Adhered RUS (As Measured)	Adhered RUS (Corrected)
c_{11} (GPa)	315.1 ± 1.2	317.8 ± 2.2	314.2 ± 2.1
c_{12} (GPa)	101.8 ± 1.4	97.4 ± 2.7	99.1 ± 2.5
c_{44} (GPa)	121.6 ± 0.2	123.8 ± 0.5	121.0 ± 0.4
B (GPa)	172.9 ± 1.0	170.9 ± 1.9	170.8 ± 1.8
ν	0.244 ± 0.003	0.235 ± 0.005	0.240 ± 0.005

was $2.0582(5)$ g yielding a density of $16.82(3)$ g/cm³, which is comparable to the literature.⁴⁰ The tungsten alloy sample exhibited isotropic elasticity based upon an elastic constant determination with the first 52 resonant frequencies (see Tables II–IV). Initial conditions were varied to confirm the reported sets of elastic constants correspond to a global minimum solution. Errors reported correspond to 2% change in the χ^2 value.¹³ Explanations for the (k, i) mode naming convention are provided in Ref. 13.

APPENDIX C: APPLICATION TO SINGLE CRYSTAL SrTiO_3

We also applied this method to a SrTiO_3 single crystal sample to show the adhered RUS approach works for anisotropic single crystals. The SrTiO_3 single crystal sample was

cut into a cylindrical shape [diameter of $2.984(5)$ mm and thickness of $3.002(5)$ mm] with the [100] crystallographic direction along the long axis of the cylinder. The mass of the sample was $0.1057(5)$ g [density of $5.04(4)$ g/cm³]. Table V provides the elastic constants determined using conventional RUS prior to the application of the adhesive, which are in good agreement with literature values (Ref. 13 and references therein). After correction according to Eq. (1), the elastic constants determined from RUS measurements where the sample has been adhered to the transducers are within the uncertainties of the conventional RUS measurements.

Next, we examine the behavior of a resonant frequency of the SrTiO_3 single crystal near the structural phase transition at ~ 105 K.⁴² For this example, we have purposefully used an excessive amount of adhesive and did not let it fully

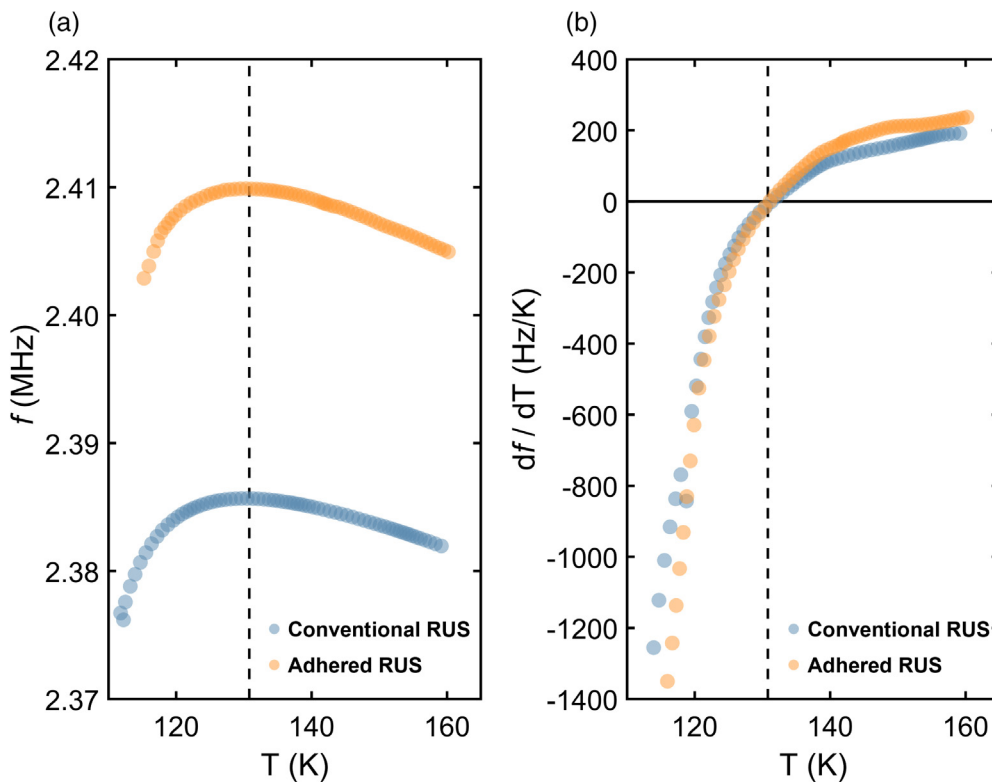


FIG. 11. (Color online) Structural phase transition in a SrTiO_3 single crystal. (a) The temperature dependence of the same resonant frequency in SrTiO_3 measured with (orange) and without (blue) an excessive amount of adhesive. (b) The onset of the softening (maximum in the temperature dependence) occurs at the same temperature, as evidenced by the temperature derivative of the same resonant frequency with (orange) and without (blue) an adhesive.

cure before cooling down the sample. Even with too much adhesive, Fig. 11 demonstrates the softening prior to the onset of the cubic-tetragonal phase transition in SrTiO_3 single crystals⁴² is apparent. The maximum in the resonant frequency also occurs at the same temperature with and without the adhesive. Though some details are affected when an excessive amount of uncured adhesive is used [e.g., curvature in Fig. 11(b)], such un-ideal circumstances still allow for the study of phase transitions. This may be important for very small samples or those which are extraordinarily unstable in magnetic fields.

APPENDIX D: CHANGES IN ULTRASONIC ATTENUATION IN ADHERED SAMPLES

Changes in the resonant frequency widths in adhered samples were investigated through measurements on a high-density tungsten alloy sample with and without adhering the sample to the ultrasonic transducers. The widths of the resonant frequencies increased in the presence of the adhesive with the extent of the increase ranging from factors of 2–15. Compared to the change in the frequencies (at most $\sim 1\%$), this change in width was large and not perturbative. There does not appear to be a strong correlation between the widths of a given resonance mode before and after the application of the adhesive. However, as shown in Fig. 12, the resonance width increase from the adhesive does seem to decrease with resonant frequency. Our simple model does not capture these effects on widths, but the frequency-dependence of the width increase in the presence of the adhesive suggests a possible connection to the spatial extent of the resonance mode vibrational pattern. Depending on the sample, such width increases may preclude quantitative analyses of internal friction, but they do not impact elastic

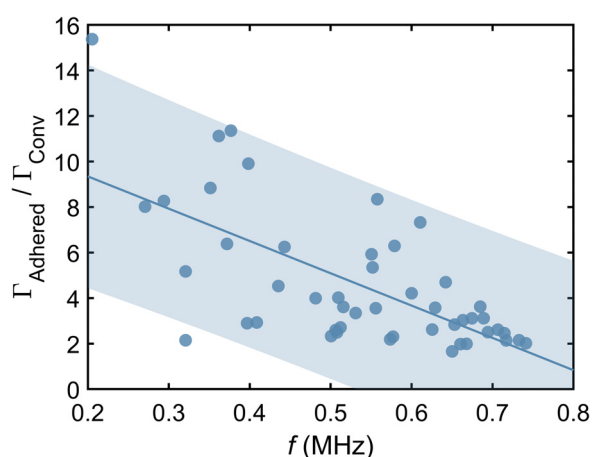


FIG. 12. (Color online) Increase in resonant frequency widths with adhesive (Γ_{Adhered}) compared to the widths measured with conventional RUS (Γ_{Conv}). The overall change in the widths ranges from a factor of 15 for the first resonant frequencies to a factor of 2 for the highest measured resonant frequencies. There is significant scatter in the data, but the increase in the resonant frequency width from the adhesive tends to decrease with frequency. Line and shaded region correspond to a linear fit to the data and the 95% confidence interval of the fit, respectively.

constant determination for strongly resonating samples because resonant frequencies only weakly depend on the resonant quality factor.¹²

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