

Detection of Alpher-Rubin attenuation and a search for nuclear acoustic resonance of surface waves in tantalum films

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The first observation of Alpher-Rubin attenuation of surface acoustic waves is reported. The surface waves were propagated on an *ST*-cut quartz substrate, and absorption occurred in a 10- μm tantalum film due to the surface-wave-driven oscillation of conduction electrons in an external 24-kG magnetic field. A scaling law is derived that relates attenuation coefficients of bulk and surface waves. Unsuccessful searches were made for nuclear acoustic resonance (NAR) of surface waves due to electric quadrupole transitions of ^{181}Ta and magnetic dipole transitions of ^1H . Theoretical estimates of the attenuation of surface waves agree with the observed Alpher-Rubin effect and indicate that the NAR signal was less than the noise.

I. INTRODUCTION

Surface acoustic waves (SAW's) are elastic waves that propagate along the free surface of a solid with acoustic energy confined to within one wavelength of the surface.¹ This wave solution was first found by Lord Rayleigh for an isotropic solid.² Piezoelectric crystals also have surface-wave modes that can be launched and detected by suitable electrode patterns.¹ The propagation velocity is $\sim 3 \times 10^5$ cm/s with frequencies ~ 100 MHz for typical electrode designs. Because of the confinement near the surface, the propagation is sensitive to surface boundary conditions. Thin films in the propagation path can be used to modify boundary conditions, causing attenuation and dispersion of the wave.

This paper reports results from an experiment that measures very small changes in attenuation (10^{-4} – 10^{-2} cm $^{-1}$) of surface acoustic waves. The surface waves were propagated on *ST*-cut quartz substrates (Euler angles $\lambda=0^\circ$, $\theta=90^\circ$, $\mu=132^\circ, 75^\circ$) and attenuation occurred due to absorption by a tantalum film on the substrate in the acoustic path. Two effects were investigated. First, nonresonant Alpher-Rubin³ attenuation of the surface wave was observed when the sample was placed in a 24-kG magnetic field. This is the first detection of Alpher-Rubin attenuation using surface waves. Second, an attempt was made to observe nuclear acoustic resonance (NAR) of ^{181}Ta and ^1H in the films.

Theories specific to SAW's were developed for the effects studied. A scaling law was derived that relates the attenuation of surface waves to the characteristics of bulk-wave absorption that are more easily measured and calculated. The measured Alpher-Rubin attenuation agrees well with the scaling-law prediction. The NAR of ^1H is much too weak to be seen with this technique, and the linewidth of the ^{181}Ta NAR is probably too great to allow detection with surface waves.

II. THEORY

A. Scaling between bulk and surface waves

The absorption of acoustic energy from bulk waves is described by the attenuation coefficient of the acoustic amplitude,^{4–6}

$$\alpha = \frac{1}{2} \frac{P_v}{P_0}, \quad (1)$$

where P_v is the power absorbed per unit volume and P_0 is the incident power per unit area (flux) in the wave. The attenuation coefficient is useful because it is independent of the amplitude of the wave. This is because both P_0 and P_v are proportional to the square of the strain. For example, in an isotropic medium,

$$P_0 = \frac{1}{2} \rho v^3 \epsilon^2, \quad (2)$$

where ρ is the mass density, v is the wave velocity, and ϵ is the strain amplitude. The power absorbed, P_v , is proportional to the square of quantum-mechanical matrix elements^{5,6} which are proportional to strain.

For a surface wave, the attenuation coefficient can be found by perturbation techniques.⁴ This results in an expression with the power flux per unit width in the denominator. Then the surface-wave attenuation must be

$$\alpha_{\text{sur}} = \frac{1}{2} \frac{P_a}{P_w}, \quad (3)$$

where P_w is the power flux per unit width and P_a is the power absorbed per unit area of the surface.

The power per unit area absorbed from the surface wave is

$$P_a = \int_{-\infty}^0 P_v dx_3, \quad (4)$$

where x_3 is the coordinate normal to the surface. The power flux per unit width of the surface wave can be calculated from the eigenvalue solution for the wave.¹

A scaling law has been developed for isotropic materials that allows attenuation results obtained for bulk waves to be applied to surface acoustic waves. This is much simpler than calculating the attenuation from first principles using perturbation techniques or Eqs. (3) and (4). The stresses at the surface, and other surface-wave properties, have been calculated as a function of the power per unit width and the frequency of the surface wave for a variety of materials.⁷ For the *ST*-cut quartz

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substrates used in these experiments, the main component of stress is

$$\sigma_0 = 2.42(P_w \omega)^{1/2}, \quad (5)$$

in cgs units, where ω is the angular frequency. The strain at the surface is $\epsilon_0 = \sigma_0/c_0$, where $c_0 = \rho v^2$ is the elastic stiffness in isotropic materials. Then,

$$P_w = 0.171 \rho^2 v^4 \epsilon_0^2 / \omega. \quad (6)$$

(σ_0 and ϵ_0 are the amplitudes.) A bulk wave with the same strain amplitude (ϵ_0) would have a power flux per unit area

$$P_a^0 = \frac{1}{2} \rho v^3 \epsilon_0^2, \quad (7)$$

and, thus, for the same strain amplitude,

$$P_w = 0.342 \frac{\rho v}{\omega} P_a^0. \quad (8)$$

The absorption of the SAW per unit area in a film on the surface is the absorption per unit volume at the surface multiplied by the thickness of the film. If the film thickness is comparable to the wavelength, then one must integrate the actual absorption over depth [Eq. (4)]. Then,

$$P_a = P_v^0 d, \quad (9)$$

where P_v^0 is the bulk absorption at the surface strain and d is either the thickness of a thin film or the effective depth of the absorption from the integration. The attenuation of the surface wave is now related to the attenuation of a bulk wave with a strain amplitude equal to the strain at the surface for the SAW:

$$\alpha_{\text{sur}} = \frac{P_v^0 d}{2P_a^0(0.342\rho v/\omega)} = 2.92 \frac{\omega d}{\rho v} \alpha, \quad (10)$$

in cgs units.

Recalling that α for the bulk is independent of the strain amplitude, Eq. (10) is generally applicable, independent of power or strain. This equation allows the use of data and theoretical calculations for bulk attenuation for the estimation of the attenuation of surface waves. However, an actual film on the surface will perturb the surface wave due to the change in elastic properties and the change in electrical boundary conditions imposed on the piezoelectric material.⁴ In addition, the elastic stiffness is a tensor for the piezoelectric substrate, not the scalar c_0 . In spite of these simplifications, Eq. (10) shows the functional relationship between bulk- and surface-wave attenuation and can be used as a scaling law.

B. Alpher-Rubin absorption

Nonresonant acoustic absorption occurs in a conductor in an external magnetic field due to the induction of oscillating magnetic fields from the motion of the conduction electrons, driven by the wave. This "Alpher-Rubin" attenuation is,³ for bulk waves

$$\alpha_{\text{AR}} = \frac{\sigma}{2\rho v c^2} \frac{\beta^2}{1+\beta^2} B^2 f(\theta), \quad (11)$$

where σ is the electrical conductivity, c is the velocity of light, B is the external magnetic field, $\beta = \omega c^2 / 4\pi\sigma v^2$, and $f(\theta) = \sin^2\theta$ for longitudinal waves and $f(\theta) = \cos^2\theta$ for transverse waves, where θ is the angle between the external magnetic field and the propagation direction of the wave. Surface waves are a specific superposition of transverse and longitudinal components that yield an oscillation amplitude that decays into the surface and all of the components of which propagate at the same velocity.¹

C. NMR absorption

In NMR the power absorbed per unit volume by a set of nuclear spins is⁸

$$P_v = \frac{n}{2I+1} \frac{(h\nu)^2}{kT} \sum_m W_{mm'}, \quad (12)$$

where n is the number density of spins, I is the total spin, ν is the frequency of the perturbation, and $W_{mm'}$ is the transition rate from the Zeeman-energy state with spin component m to the state with component m' . The rate is given by⁸

$$W_{mm'} = \frac{\pi^2}{h^2} |\langle m | H | m' \rangle|^2 g(\nu), \quad (13)$$

where H is the time-varying perturbation Hamiltonian at frequency ν and $g(\nu)$ is a line-shape factor, the integral over ν of which is unity. Again, in principle, Eqs. (3), (4) (12), and (13) can be used to calculate α_{sur} . However, α for bulk acoustic NMR can also be scaled with Eq. (10).

Magnetic dipole NMR absorption occurs through the interaction of the oscillating (perturbing) magnetic fields generated as described above (in the discussion of Alpher-Rubin attenuation) with the dipole moments of the nuclei. The attenuation coefficient for bulk waves is⁹

$$\alpha = \frac{\omega B^2 \sin^2\theta \cos^2\theta}{2\rho v^3} \left[\frac{1-\beta^2}{(1+\beta^2)^2} \right] \left[\chi'' - \left[\frac{2\beta}{1-\beta^2} \right] \chi' \right], \quad (14)$$

where χ' is the in-phase magnetic susceptibility and χ'' is the out-of-phase magnetic susceptibility of the material. The matrix elements, which are generally not calculable, are contained in the susceptibilities. The magnetic dipole transition selection rules require that $\Delta m = \pm 1$, and the frequency of the perturbation at which resonance occurs will be $\omega = \gamma B$, where γ is the gyromagnetic ratio for the nucleus of interest.

Electric quadrupole NMR absorption occurs because the oscillating strain in the lattice produces a perturbing electric field gradient that interacts with the electric quadrupole moments of the nuclei. The transitions are between Zeeman-energy levels. Electric quadrupole matrix elements are calculable because the perturbation can be expanded in spherical harmonics, which are the angular-momentum (energy) eigenfunctions.¹⁰ The electric quadrupole transition selection rules allow $\Delta m = \pm 1$

or ± 2 . The resonant frequency of the $\Delta m = \pm 2$ transitions is twice the value for $\Delta m = \pm 1$. The attenuation for $\Delta m = \pm 2$ for transverse bulk waves is¹¹ (note that Ref. 11 incorrectly has v^2 in the denominator)

$$\alpha = \frac{\pi^2}{4} \frac{\sum F(I)^2}{(2I)^2(2I-1)^2(2I+1)^2} \frac{nv^2e^2Q^2S^2}{\rho v^3kT} g(v)\sin^2\theta, \quad (15)$$

where $F(I) = [(I \mp m)(I \mp m - 1)(I \pm m + 1)(I \pm m + 2)]^{1/2}$ and the sum is over all the allowed transitions, e is the electron charge, Q is the scalar quadrupole moment of the nucleus, k is Boltzmann's constant, T is the absolute temperature, and S is an element of the tensor that relates the strain in the lattice to the electric field gradient. The exact element S that is used depends on the propagation direction relative to the crystal axes; an additional numerical factor ($\sim \frac{1}{2}$) may be required in Eq. (15) for different propagation directions.

III. EXPERIMENT

Figure 1 shows a schematic sketch of the sample design for the propagation of a surface acoustic wave through a film on the surface of a piezoelectric substrate. The surface-wave device is a standard SAW delay line. The surface wave is launched by an interdigital electrode pattern at one end and detected by an identical transducer at the other end. The surface-wave velocity is the eigenvalue for the superposition of wave modes that decay into the interior.¹ Thus, the electrode spacing is chosen so that the applied oscillating voltage produces a periodic strain in the piezoelectric substrate with a wavelength that is the surface-wave velocity divided by the applied frequency. Since the detecting transducer couples most efficiently to waves of the correct wavelength, this delay line is a narrow-bandpass filter. The frequency of operation is thus restricted.

The SAW devices were made by Crystal Technology¹² to the author's designs. The substrate used was ST-cut quartz. The tantalum films were deposited between the transducers by electron-beam evaporation. The tantalum would not adhere to the optical-quality surface of the quartz, so an underlayer of chromium, $\sim 0.2 \mu\text{m}$ thick, was used to provide adhesion for the Ta film.

The attenuation of the surface wave in this sample is detected by a transmission marginal oscillator.¹³ Figure 2 shows a block diagram of the basic circuit. The sample

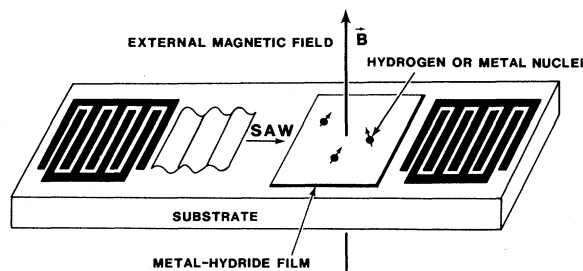


FIG. 1. Schematic sketch of the surface-wave sample.

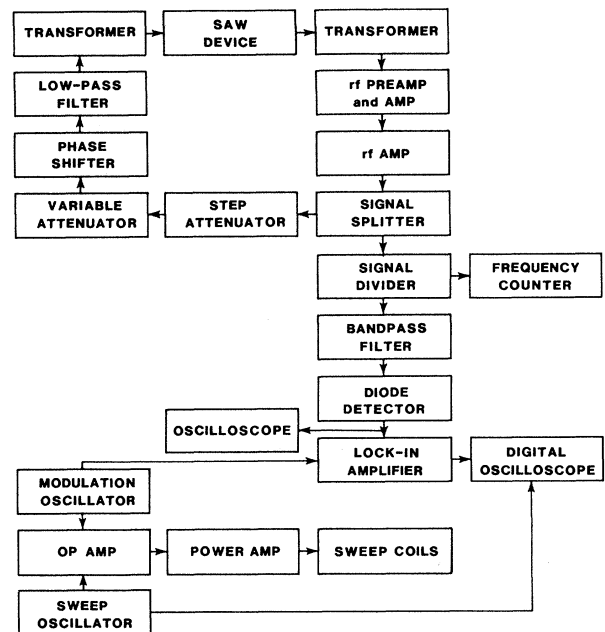


FIG. 2. Schematic of the marginal oscillator.

is placed in the feedback loop of an oscillator which uses radiofrequency amplifiers. Stable oscillation occurs only when the phase shift around the loop is a multiple of 2π and the product of the amplifier gain and the total loss is unity. The gain of any amplifier decreases slightly with increasing signal level and ultimately saturates. The attenuators are adjusted to result in a total loss that requires a small oscillation amplitude to give the required gain for oscillation. Thus, a very small change in attenuation in the sample will be compensated by a small change in gain of the amplifier to maintain oscillation. This will change the signal level a measurable amount.

A portion of the oscillation signal is split from the feedback loop and detected with a diode, yielding the oscillation level. The external magnetic field is modulated and phase-sensitive detection of the oscillation level is performed with a lock-in amplifier to reduce random noise. The external field is also swept, and the lock-in output for successive sweeps can be averaged by a digital oscilloscope to further reduce noise.

The change in transmitted power of an electrical circuit is usually expressed in decibels, and the power change in dB, ΔP , is

$$\Delta P = -\alpha_{\text{sur}} l 20 \log_{10} e, \quad (16)$$

where l is the acoustic path length. For very small changes in acoustic loss (i.e., to first order), the change in detector output, ΔV , is proportional to the change in power due to the sample:

$$\Delta V = -C \Delta P, \quad (17)$$

where C is the sensitivity (e.g., mV/dB) of the oscillator. The oscillator was calibrated at various oscillation levels

by inserting 0.1-dB attenuators into the feedback loop and noting the change in oscillation level. This change in attenuation is very large compared to the change expected for effects in the film, and the nonlinearity of the response was recognized by averaging the beginning and ending oscillation levels (before and after the 0.1-dB insertion) as the oscillation level appropriate to the sensitivity observed. The lock-in amplifier detects the root-mean-square signal due to the sinusoidal modulation:

$$s = \frac{1}{\sqrt{2}} \frac{\partial V}{\partial B} \delta B, \quad (18)$$

where δB is the amplitude of the modulation. Thus, the lock-in signal is

$$s = \frac{20}{\sqrt{2}} C I \log_{10}(e) \frac{\partial \alpha_{\text{sur}}}{\partial B} \delta B. \quad (19)$$

Each sample was attached to a small circuit board and very fine gold wires were bonded to the transducers to provide electrical connection. A sample was placed in a probe to position it in a 24-kG electromagnet. The probe could be rotated to provide different angles between the SAW-propagation direction and the magnetic field. Magnetic field sweep and modulation could be provided at amplitudes up to $\delta B \sim 65$ G.

IV. RESULTS

A. Alpher-Rubin absorption

Alpher-Rubin attenuation was detected in a 10- μm tantalum film with an oscillation frequency of 24 MHz and an acoustic path length of 0.432 cm. The Alpher-Rubin attenuation signal was acquired on the digital oscilloscope and the average signal s and the standard deviation Δs were calculated over an acquisition period of ~ 16 s. The lock-in-amplifier integration time constant was 1 s and the modulation amplitude was $\delta B = 60$ G. The sensitivity was $C = 10$ mV/dB, with an average signal-to-noise ratio of $s/\Delta s = 28$. A typical signal was $s = 2.17 \mu\text{V}$ with $\Delta s = 0.10 \mu\text{V}$ at $\theta = \pi/4$.

Figure 3 shows the observed signal s as a function of the angle θ between the SAW-propagation direction and

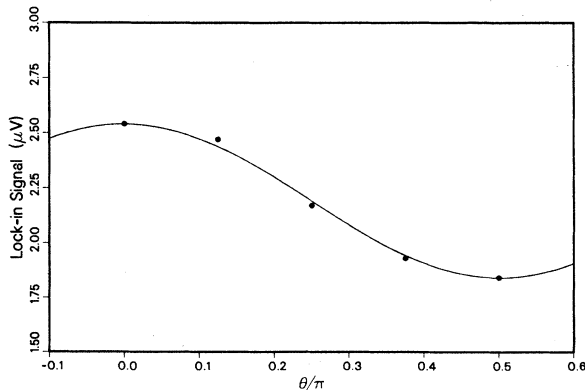


FIG. 3. Observed Alpher-Rubin attenuation signal as a function of the angle between the wave-propagation direction and the external magnetic field. The error of each data point is $\sim 0.08 \mu\text{V}$.

the external magnetic field. The curve is $s = 1.84 \sin^2\theta + 2.54 \cos^2\theta \mu\text{V}$, which shows that the SAW consists of both transverse and longitudinal components [Eq. (11)].

The predicted Alpher-Rubin attenuation for a surface wave in the pure Ta film at 23.6 kG and 24 MHz is $\alpha_{\text{AR}}^{\text{sur}} = 1.5 \times 10^{-2} \text{ cm}^{-1}$ [Eqs. (10) and (11)]. (Note that $\rho = 2.2 \text{ g/cm}^3$; the mass density is that of quartz since the density enters through the elastic properties of the substrate.) From Eq. (19), the lock-in signal should be $s_{\text{AR}} = 2.0 \times 10^{-6} \text{ V}$, very close to the magnitude observed. From Eqs. (11) and (19), we see that the lock-in signal should be proportional to B . Measurements at $B = 0, 10,$ and 24 kG showed strict linearity.

B. Magnetic dipole NMR

Alekseev¹⁴ first proposed using surface waves to stimulate NMR. At the time, however, SAW technology did not exist to make the samples described above. Alekseev calculated the ratio of attenuation coefficients for SAW and bulk-wave electric quadrupole resonance by integrating the matrix elements over depth using the Rayleigh surface-wave strains. The results were $\alpha_{\text{sur}} = 2.41\alpha$ in MgO and $\alpha_{\text{sur}} = 4.04\alpha$ in RbBr. Since the matrix elements are proportional to the square of the strain, the effective depth of the surface wave is $d \approx \int \epsilon^2 dx_3 / \epsilon_0^2$. This is $d \sim 0.3\lambda$, where λ is the wavelength.¹⁵ Then, using Eq. (10), $\alpha_{\text{sur}} = 1.58\alpha$ for MgO ($\rho = 3.58 \text{ g/cm}^3$) and $\alpha_{\text{sur}} = 1.66\alpha$ for RbBr ($\rho = 3.35 \text{ g/cm}^3$). These results are in reasonable agreement, considering the scalar lowest-order derivation of Eq. (10) and the fact that Eq. (10) uses the SAW strain for *ST*-cut quartz, not MgO or RbBr. Since the NMR attenuation of surface waves appeared to be similar to the NMR attenuation of bulk acoustic waves, and nuclear acoustic resonance using bulk acoustic waves is a well-developed technique,^{5,6} the author attempted to observe nuclear acoustic resonance using surface waves.

An attempt was made to observe the resonance of ^1H in a 5- μm -thick film of tantalum that was loaded with hydrogen to form Ta_2H . This film was deposited on a SAW device designed to operate at 98.6 MHz, the resonant frequency of ^1H at ~ 24 kG. The acoustic path length was 0.432 cm. No attenuation, either resonant or non-resonant, was observed. The calibration of the oscillator was $C = 142.5$ mV/dB and the noise in the signal was $\Delta s \sim 5 \mu\text{V}$. The search for the resonant signal was performed at various lock-in-amplifier settings (integration time constant, etc.) and various modulation amplitudes by varying the average external magnetic field about the resonant value and observing many sweeps of the external field.

Assume that the NMR resonance line is roughly triangular. Then, the derivative is $\partial\alpha/\partial B \approx \alpha_m/W$, where α_m is the maximum absorption strength of the line and W is the full width at half maximum. The attenuation is $\alpha = \alpha_0 g(B)$, where $g(B)$ is the line-shape factor, the integral of which over B is unity and $g(\nu) = 2\pi g(B)/\gamma$. Then, $\alpha_m = \alpha_0 g(B_m)$, where B_m is the field at the max-

imum of $g(B)$ and $g(B_m) = 1/W$ since the integral is unity. Thus, $\partial\alpha/\partial B \approx \alpha_0/W^2$ and the lock-in signal is

$$s = \frac{20}{\sqrt{2}} Cl \log_{10}(e) \frac{\alpha_0^{\text{sur}}}{W^2} \delta B, \quad (20)$$

where α_0^{sur} is the total strength of the line in the surface-wave case.

NAR of hydrogen using bulk acoustic waves has been reported only once, for $\text{NbH}_{0.026}$.⁹ The observed attenuation was $\alpha = 2 \times 10^{-8} \text{ cm}^{-1}$ at 210 MHz and 49 kG. This is a very weak effect. The linewidth was 0.4 G, and the resonance was observed by averaging 192 sweeps at 72 s per sweep. The total line strength was therefore $\alpha_0 = 8 \times 10^{-9} \text{ cm}^{-1}$. The reported susceptibility was $\chi'' + 0.3\chi'$, or $\beta = 6.8$ [Eq. (14)].

Since both Ta and Nb are bcc group-V transition metals, assume that χ' and χ'' are similar. Then scale the observed value of α_0 in Nb to give the expected signal s in the present experiment. For Ta, $\sigma = 5.76 \times 10^{16} \text{ s}^{-1}$, $\rho = 16.65 \text{ g/cm}^3$, and $\beta = 2.08$ at $\omega = 6.2 \times 10^8 \text{ s}^{-1}$. However, the hydrogen in the film lowers the conductivity, and a conductivity of $\sigma = 6.7 \times 10^{15} \text{ s}^{-1}$ was measured for the film on the SAW device, giving $\beta = 74$. Scaling with Eq. (14) to the film conditions gives $\alpha_0 = 8 \times 10^{-11} \text{ cm}^{-1}$. Equation (10) then gives $\alpha_0^{\text{sur}} = 1.1 \times 10^{-10} \text{ cm}^{-1}$. Using Eq. (20), with $W \sim \delta B \sim 1 \text{ G}$, the expected lock-in signal is $s \sim 4 \times 10^{-11} \text{ V}$. This is a factor of 10^5 less than the observed noise for one sweep.

In addition to the search for nuclear acoustic resonance, this sample was used to search for Alpher-Rubin absorption. The Alpher-Rubin nonresonant absorption was not detected in the $5\text{-}\mu\text{m}$ Ta_2H film. The lock-in signal is

$$s_{\text{AR}} = \frac{20}{\sqrt{2}} Cl \log_{10}(e) \frac{2\alpha_{\text{AR}}^{\text{sur}} \delta B}{B}, \quad (21)$$

where $\alpha_{\text{AR}}^{\text{sur}}$ is the Alpher-Rubin attenuation of the surface wave. From Eq. (11), $\alpha_{\text{AR}} = 3.2 \times 10^{-3} \text{ cm}^{-1}$. From Eq. (10), $\alpha_{\text{AR}}^{\text{sur}} = 4.5 \times 10^{-3} \text{ cm}^{-1}$. Then, from Eq. (21), $s_{\text{AR}} = 1.4 \times 10^{-7} \text{ V}$. This is a factor of ~ 40 less than the noise for one sweep.

C. Electric quadrupole NMR

Another attempt was made to detect attenuation of surface waves due to NMR. The ^{181}Ta nucleus has one of the largest electric quadrupole moments, $Q = 3.9 \text{ b}$. Thus, an attempt was made to observe attenuation due to a pure $10\text{-}\mu\text{m}$ tantalum film at the frequency of the

$\Delta m = \pm 2$ transitions for ^{181}Ta , $\nu = 24 \text{ MHz}$ in the 24-kG magnet. No nuclear acoustic resonance was detected, but the Alpher-Rubin attenuation was observed as described above.

From Eq. (15), the resonant quadrupole attenuation should be about $\alpha = 4.0 \times 10^{-4} \text{ g}(B) \text{ cm}^{-1}$ in the film. [$I = \frac{7}{2}$ for ^{181}Ta , $\sum F(I)^2 = 1008$,⁵ $|S_{44}| = 2.11 \times 10^{16} \text{ statcoulombs/cm}^3$,¹⁶ and $\rho = 2.2 \text{ g/cm}^3$.] Then, $\alpha_0^{\text{sur}} = 2.7 \times 10^{-4} \text{ cm}^{-1}$ [Eq. (10)] and the expected signal is $7.2 \times 10^{-6} \delta B/W^2 \text{ V}$. The largest practical modulation amplitude is $\delta B/W \sim 0.3$, or the phase-sensitive detection will integrate over the line and it will not be detected. Then $s \approx 2.2 \times 10^{-6}/W \text{ V}$.

The linewidth of the quadrupole transition is fairly broad, $W \approx 40 \text{ G}$ at $T = 4.2 \text{ K}$.¹⁷ Any deviation from cubic symmetry further broadens the line because of the electric field gradients induced in the lattice.¹⁸ For example, small amounts of hydrogen in tantalum strain the lattice and broaden the line, $W = 250\text{--}300 \text{ G}$ for $\text{TaH}_{0.00488}$.¹⁷ Films typically have very high stresses and strains¹⁹ and the resonance line could conceivably be very broad. Even a narrow line of $W = 40 \text{ G}$ would give a signal of only $s \approx 0.05 \mu\text{V}$, comparable to the lowest noise achieved.

V. SUMMARY

This paper describes the study of very small changes in propagation of surface acoustic waves in films. Nonresonant Alpher-Rubin attenuation was detected with an attenuation coefficient of the surface wave of only $\alpha = 1.5 \times 10^{-2} \text{ cm}$. A scaling law [Eq. (10)] will allow the use of bulk acoustic data to estimate the magnitude of physical effects on surface-wave propagation. Searches for nuclear acoustic resonance were unsuccessful and theoretical considerations show that the NAR effects were less than the noise.

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