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Detection of Shorter-Than-Skin-Depth Acoustic Pulses in a Metal Film via Transient Reflectivity

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Abstract. Short acoustic pulses are generated in SrRuO\textsubscript{3} transducers and detected at the surface of gold films by transient reflectivity. Contrary to expectations, acoustic pulses that are shorter than the optical skin depth of gold are resolved. A comparison of gold detection films that were grown under different deposition conditions demonstrates that the microstructure of a detection film can impact the shape of the detected signal.

Keywords: Laser Ultrasonics, Thin Film Morphology

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INTRODUCTION

Optical generation and detection of ultrashort acoustic pulses is often performed with thin metal film transducers. The detection typically relies on the modulation of the optical constants by acoustic strain and is accomplished by measuring the transient reflectivity of the metal surface using a variably delayed probe pulse\textsuperscript{1}. Because the probe light penetrates into the material over a finite distance, it is intuitive to suggest that the ability to resolve a short acoustic pulse by transient reflectivity is limited by the optical skin depth\textsuperscript{2}. Consequently, aluminum, which has a short skin depth of \textasciitilde 7 nm in the visible range, is typically considered the material of choice for transducer films in picosecond ultrasonics\textsuperscript{3,4}. Recently Mante et al.\textsuperscript{5} demonstrated the detection of acoustic pulses significantly shorter than the optical skin depth in semiconductor InP. In this report, we show that the detection of sub-skin-depth acoustic pulses is also possible in a common metal such as gold. We show that the transient reflectivity response of any strongly absorbing material to a shorter-than-skin-depth acoustic pulse can be decomposed into two terms: a slow response whose shape is entirely determined by the optical constants of the medium and is independent of the acoustic pulse duration, and a term containing a sharp step that represents the integral of the acoustic strain profile. The superposition of the two components leads to an intricate shape of the observed waveforms. In this experiment, we use gold films grown under two sets of deposition conditions for the detection of acoustic pulses generated in very thin (4-12 nm) layers of SrRuO\textsubscript{3} (SRO). We will show that the detection bandwidth of the transient reflectivity method is limited not by the skin depth but rather by the microstructure and the roughness of metal films.

THEORY

Consider a longitudinal acoustic pulse incident on a free surface of a medium occupying a semi-infinite half-space \( z > 0 \). Assume, initially, that the pulse is infinitely short and the acoustic strain profile can be represented by a \( \delta \)-function,

\[
\eta_{33}(z, t) = \begin{cases} 
\delta(z + vt), & t < 0 \\
-\delta(z - vt), & t > 0 
\end{cases}
\]  

(1)

where \( v \) is the longitudinal acoustic velocity. The acoustic strain changes sign upon reflection from the free surface at \( t = 0 \).

The change in reflectivity caused by acoustic strain is given by\textsuperscript{5}
where $f(z)$ is the sensitivity function defined in Thomsen, et al. For the purposes of our analysis, it is convenient to represent the sensitivity function in the following form,

$$
\Delta R(t) = \int_0^\infty f(z) \eta_{33}(z,t)\,dz
$$

(2)

where $f(z)$ is the sensitivity function defined in Thomsen, et al. For the purposes of our analysis, it is convenient to represent the sensitivity function in the following form,

$$
f(z) = A \sin(2k_0 nz) e^{-nz} + B \cos(2k_0 nz) e^{-nz}$$

$$A = C \frac{\partial n}{\partial \eta_{33}} + D \frac{\partial \kappa}{\partial \eta_{33}}$$

$$B = D \frac{\partial n}{\partial \eta_{33}} - C \frac{\partial \kappa}{\partial \eta_{33}}$$

$$C = \frac{8k_0 n(n^2 + \kappa^2 - 1)}{[(n + 1)^2 + \kappa^2]^3}$$

$$D = \frac{8k_0 \kappa(n^2 + \kappa^2 + 1)}{[(n + 1)^2 + \kappa^2]^3}$$

(3)

where $k_0 = 2\pi/\lambda$ is the optical wavenumber in vacuum, $n$ and $k$ are the real and imaginary parts of the refractive index, and $\zeta = \lambda/4\pi\kappa$ is the optical absorption depth.

The transient reflectivity response to the $\delta$-pulse defined by Eq. (1) is given by:

$$G(t) = -A \sin(2k_0 nvt) e^{-|v|\zeta} - B \cos(2k_0 nvt) e^{-|v|\zeta}$$

(4)

In the case of a strongly absorbing medium $k \gg n$, Eq. (4) can be simplified as follows:

$$G(t) = -2Ak_0 nvt e^{-|v|\zeta} - B \cos(2k_0 nvt) e^{-|v|\zeta}$$

(5)

Fig. 1 shows the components of the response which correspond to the two terms in Eq. (4), as well as examples of a total response corresponding to $B/A = 1/3$ and $B/A = -1/3$ for $n = 1.67$ and $k = 1.94$, the latter values corresponding to gold at 393 nm. When $A$ and $B$ have opposite signs, the response is characterized by a particularly intricate shape. For an incoming acoustic pulse with an arbitrary profile of $\eta_{33} = F(z/v + t)$, the response will be given by a convolution,

$$\Delta R(t) = v \int_{-\infty}^{\infty} F(t' - t) G(t') dt'$$

(6)

where the factor $v$ originates from the identity $\delta(z/v + t) = v \delta(z + vt)$. The first term in Eq. (4) is a smooth function; after a convolution with a short pulse it yields the same smooth function, which is entirely determined by the optical constants and the speed of sound of the medium and contains no information about the strain profile. The second term in Eq. (4) contains a sharp step, which, after a convolution with a short pulse, will yield an integral of the pulse profile.
FIGURE 1. The transient reflectivity response corresponding to the (a) first and (b) second term of Eq. (4); simulated responses for \( n = 1.67, k = 1.94, \) and for a ratio (c) \( B/A = 1/3 \) and (d) \( B/A = -1/3. \)

It is instructive to consider the time-derivative of the transient reflectivity response,

\[
\frac{dR}{dt} = -2vnBF(t) + \left(-\frac{2k_0nA - B}{\zeta}\right)\int_{-\infty}^{\infty} F(t - t') \cos(2k_0nv't)e^{-\frac{\zeta}{2}t'}dt' + \left(\frac{2k_0nB + A}{\zeta}\right)\int_{-\infty}^{\infty} F(t - t') \sin(2k_0nv't)e^{-\frac{\zeta}{2}t'}dt' \tag{7}
\]

For a short acoustic pulse with duration \( t \), the second term in Eq. (7) is proportional to \( t \) and the third term is proportional to \( r^2 \). Consequently, in the short-pulse limit the first term is dominant and the derivative of the reflectivity response yields the acoustic strain profile.

It is also instructive to consider the long-pulse limit. If \( F(t) \) is a slowly varying function on the time scale of \( \zeta/v \), then a first-order Taylor expansion of Eq. (6) yields:

\[
\Delta R(t) = -\frac{2\zeta^2}{v\left(1 + 4k_0^2n^2\zeta^2\right)^2} \left[4k_0n\zeta A + \left(1 - 4k_0^2n^2\zeta^2\right)B\right] \frac{dF}{dt} \tag{8}
\]

In this case both terms in \( G(t) \) yield a signal that is proportional to the derivative of the acoustic pulse profile. This is noteworthy: a long pulse is differentiated by the detection process, whereas a short pulse is integrated.

We see that in order to detect a short acoustic pulse, one needs a material and probe wavelength combination that yields a large value of \( B \). It is easy to see that \( B \) vanishes for a weakly absorbing medium with vanishing \( k \), hence a strongly absorbing material is desired. On the other hand, \( B \) also vanishes in the limit \( k \gg n \). Thus a smaller skin depth should not necessarily be preferred. Because detailed data on the wavelength dependence of the photoelastic constants \( \partial n/\partial \eta_{33} \) and \( \partial k/\partial \eta_{33} \) are typically lacking, one has to resort to trial-and-error in search for the best material / wavelength combination. As is shown below, gold works well at 400 nm yielding \( B/A = -0.71 \pm 0.08 \).
EXPERIMENTAL

The experimental geometry is pictured in Fig. 2(a). The samples consisted of layered SRO/gold structures grown on SrTiO$_3$ (STO) substrates. SRO layers with thicknesses from 4 to 12 nm were grown using 90 degree off-axis RF magnetron sputtering at a rate of 1 nm/min. Polycrystalline gold films with primarily (111) orientation and a thickness of ~200 nm were grown using magnetron sputtering, and the sputtering conditions were varied to produce two sets of gold films with quantitatively different material properties. From high-resolution scanning electron microscope (SEM) images of the two gold films, it is evident that the first set of gold films has an average grain diameter of ~400 nm (Fig. 3(a)) and the second set of films has an average grain diameter of ~40 nm (Fig. 3(b)).

![Figure 2](image1.png)

**FIGURE 2.** (a) Schematic diagram of the sample structure and the experimental geometry; (b) transient reflectivity detected at the free surface of the gold layer.

![Figure 3](image2.png)

**FIGURE 3.** (a) SEM images collected at the free surface of (a) gold film #1 and (b) gold film #2.

A Mira Ti-sapphire oscillator with a RegA regenerative amplifier was employed to generate laser pulses with a central wavelength of 784 nm and duration of 300 fs. The laser output was split into separate pump and probe beams and the probe beam was frequency-doubled to a wavelength of 393 nm. Illumination of the SRO transducer layer with the pump pulse launched an acoustic strain pulse through the gold layer, and the strain pulse was detected at the gold/air interface through the transient reflectivity of the variably delayed probe pulse.
RESULTS AND ANALYSIS

The reflectivity signal is depicted in Fig. 2(b). The sharp spike at 0 ps and subsequent peaks at 18 ps and 35 ps arise from fast diffusion of hot electrons across the gold layer that is initiated by the pump pulse and its reflections within the 1 mm thick SrTiO$_3$ substrate. The slow rise in the background signal is due to a temperature increase at the gold surface from the thermal equilibration of the hot electrons with the lattice. The signal at 60 ps marks the arrival of the acoustic strain pulse at the gold surface. It contains a fast, 2-ps component, which corresponds to the integral of the strain pulse profile, in addition to a slowly varying component, which arises from the interaction of the strain pulse with the light that penetrates into the film. Based on the optical skin depth of gold ($\zeta = 16.4$ nm), we would not expect to resolve an acoustic pulse shorter than ~4 ps ($v_{Au,(111)} = 4.05$ nm/ps). The experimental response is similar in shape to the simulated response in Fig. 1(d).

A comparison of the transient reflectivity responses of the two gold films and their time-derivatives is shown in Fig. 4(a-d). The overall signal strength is similar, but the magnitude of the short central feature is significantly smaller in gold film #2. The Fourier transform spectra of the time-derivatives indicate that the detection bandwidth of gold film #1 extends to ~350 GHz, and the detection bandwidth of gold film #2 extends to ~300 GHz. Applying a shorter strain pulse by exciting a thinner transducer layer extends the bandwidth of gold film #1 to ~450 GHz. These results indicate that the bandwidth is limited by the detection mechanism or by acoustic broadening within the film.

FIGURE 4. (a) and (b) Transient reflectivity response of gold film #1 and gold film #2, respectively, to an acoustic strain pulse generated in a 12 nm SRO transducer; (c) and (d) the time derivative of the response and the fits to Eq. (7) for gold film #1 and #2, respectively.

To estimate the ratio $B/A$ for the two gold films, we fit the derivative of the reflectivity to the functional form described by Eq. (7) using a simulated strain pulse. Because the low acoustic impedance mismatch between STO and SRO suppresses the reflected wave ($Z_{SRO} = 41$ MRayls and $Z_{STO} = 40$ MRayls), the temporal profile of the simulated strain pulse was approximated as a rectangle with a width of $d_{SRO}/v_{SRO} = 1.90$ ps for a 12 nm SRO transducer, where $d_{SRO}$ is the thickness of the SRO layer and $v_{SRO}$ is the acoustic velocity of SRO. The simulated strain pulse was convoluted with a Gaussian profile with a width of $\tau_{pulse}$ to account for acoustic broadening as the pulse traversed the gold film. The fit parameters were the ratio $B/A$ and the width of the Gaussian $\tau_{pulse}$.

We fit the time derivative of the transient reflectivity response of gold films #1 and #2 to acoustic strain pulses generated in 4-nm and 12-nm thick SRO transducers. Examples of the fitted forms for are pictured in Fig. 4(c,d). For a strain pulse generated in a 12-nm thick SRO transducer, the first gold film gave a ratio $B/A = 0.71 \pm 0.08$ and a
Gaussian broadening of $\tau_{\text{pulse}} = 0.96 \pm 0.05$ ps, and the second gold film gave a ratio $B/A = -0.60 \pm 0.08$ and a Gaussian broadening of $\tau_{\text{pulse}} = 1.50 \pm 0.05$ ps. The fits indicate that the difference in the shape of the acoustic signal between the two films arises primarily from additional acoustic broadening in the second film.

Acoustic pulse broadening in polycrystalline thin films may be caused by acoustic damping in the material, surface roughness, variations in grain orientation, or grain-boundary scattering. For broadening that arises exclusively from surface roughness, the Gaussian width can be related to the root mean square ($\sigma_{\text{rms}}$) surface roughness of the film $\tau_{\text{pulse}} = \sigma_{\text{rms}}/\sqrt{A}$. AFM analysis indicates that gold film #1 ($\sigma_{\text{rms}} = 1.28$ nm) actually has a slightly higher surface roughness than gold film #2 ($\sigma_{\text{rms}} = 1.14$ nm); this amount of roughness would result in broadening of $\sim 0.3$ ps, which is smaller than our estimated broadening. Therefore, surface roughness variability between the two films is unlikely to be the cause of additional acoustic broadening in gold film #2. In addition, orientation analysis by electron backscatter microscopy shows that there is a greater variation in grain orientation in gold film #1 than in gold film #2. However, the difference in sample morphology, evidenced by Fig. (3), is still likely the source of the additional acoustic broadening witnessed in gold film #2.

CONCLUSION

Our results demonstrate that transient reflectivity can be used to detect shorter-than-skin depth acoustic pulses in common metals such as gold. The upper limit on the bandwidth is not determined by the optical skin depth of the material but on the material characteristics of the detection film such as microstructure and surface roughness. We have shown that in polycrystalline gold, the shape of the transient reflectivity signal depends on the grain size; therefore, attention must be paid to the deposition parameters when preparing metal films as high-frequency photo-acoustic transducers.

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