Detection of hidden gratings using light and sound

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Prior to experiments on samples with buried gratings, we performed a set of experiments to obtain a basic understanding of the acoustic wave generation, propagation, and detection in metals. In this chapter, the measurements and calculations of transient-grating pump-probe experiments on flat metal layers are discussed. We show that grating-shaped acoustic waves can be generated in Au, Ni and W layers, and also that the presence of the acoustic echo can be detected by diffracted light from the grating-shaped acoustic wave.

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5. Acoustic waves in flat metal and dielectric layers

5.1 Introduction

We performed femtosecond transient-grating pump-probe experiments on flat metal layers deposited on a glass substrate. The aim of these experiments is to see if it is possible to launch and detect acoustic echoes in the metal layers using our transient-grating pump-probe setup. Although in the past, various groups have shown the generation and detection of acoustic waves in metal layers, most of these experiments were performed using a pump-probe setup in reflection or transmission geometries [29,30,38,40,42,86,88–121]. In a typical pump-probe setup, the pump pulse launches an acoustic wave, and the change in reflectivity of the metal due to the acoustic wave is measured using the probe pulse. However, in our experiments, we launch a grating-shaped acoustic wave using two interfering pump pulses and detect diffraction from the acoustic echoes using a time-delayed probe pulse.

In this chapter, we show the results of the transient-grating pump-probe experiments on flat metal layers. The first set of measurements on flat Au layers having different thicknesses shows that our transient-grating pump-probe setup is capable of generating and detecting acoustic waves. The measurements on a 500 nm thick Au layer, where we pump from the air-metal side and probe from the air-metal side and from the glass-metal side, provide information on the various effects that determine the temporal shape of the diffraction signal, such as electron energy diffusion and the thermo-optic effects. We also perform measurements on flat Ni and W layers that show that the frequency of the acoustic waves generated in Ni and W is much higher than the one generated in the Au layer. We demonstrate that a thin layer of Ni can be used as an acoustic transducer to launch a high-frequency acoustic wave in Au. To understand how the acoustic waves propagate through a sample consisting of a metal and a dielectric, we also performed measurements on a sample that has a thin layer of SiO$_x$ sandwiched between two Au layers.
5.2 Experimental setup

We use the same transient-grating setup as the one described in Chapter 4. In the experiments shown here, we have a transient-grating period of approximately 6 µm. The spatially-periodic interference pattern excites the metal layer resulting in spatially periodic heating of the sample surface, thus launching a grating-shaped acoustic wave. The grating-shaped acoustic wave propagates through the metal layer, reflects from the metal-substrate interface, and returns to the top surface. The weaker 800 nm pulse is used as the probe and is delayed with respect to the 400 pump pulses by a mechanical delay line, which increases the optical path length. The probe pulse diffracts from the grating-shaped displacement of the metal atoms or from the spatially periodic change in the refractive index of the metal surface induced by the acoustic waves. The diffracted probe pulse is recorded by a silicon photodiode placed at the position where we expect the first-order diffracted beam. The diffracted probe signal recorded by the detector when the chopper blocks the pump beam, is subtracted from the diffracted probe signal when the pump beam is transmitted by the chopper, and plotted as a function of the pump-probe delay. The signal recorded by the detector in the absence of the pump pulse is from the probe light scattered by the surface roughness of the sample. Depending on the experiment, we probe from the air-metal side or from the glass substrate side. In the latter case, the probe pulse passes through the glass substrate and probes the metal near the glass-metal interface. The pump beam has a spot size of 1.5 mm on the sample, and the pump pulse energy ranges from 15 µJ to 20 µJ depending on the experiment. The probe pulse energy was kept constant at 1 µJ.

5.3 Results and discussion

5.3.1 Acoustic waves in Au

Our first experiment was to launch and detect grating-shaped acoustic waves in a flat metal layer (schematic in Figure 5.1 (a)). We chose Au for the initial experiments because Au is one of the most well-studied metals. In Figure 5.1 (b), we plot the measured diffraction signal as a function
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Figure 5.1: (a) Schematic of the experiment. (b) Measured diffracted signal as a function of pump-probe delay for a 100 nm, 200 nm and 500 nm Au layer on a glass substrate. The plots are vertically offset for clarity. The black curves are the simulated diffracted signals.

of pump-probe delay for 100 nm, 200 nm, and 500 nm thick flat Au layers on a glass substrate. In all the measurements, the diffraction signal increases just after the optical excitation with the pump pulses and decreases again within the first 10 ps. This signal is due to the absorption of the optical energy by the electron gas, which then cools on a time-scale of $< 10$ ps by heating the lattice (see Chapter 4), until the electron gas temperature and lattice temperature are the same.

For the 100 nm Au sample, we see an oscillatory signal with peaks at $33 \pm 3$ ps, $89 \pm 3$ ps, and $145 \pm 3$ ps. The time separation between the peaks is approximately 56 ps, which is close to the expected round trip time of an acoustic wave inside a 100 nm thick Au layer, assuming an acoustic velocity of 3200 m/s in Au [30]. When the thickness of the Au layer is doubled to 200 nm, the time separation between the peaks also doubles to about 111-112 ps. Similarly, for the 500 nm thick layer, the observed round trip time is close to 280 ps. Interestingly, for measurements on all three samples, the first peak is at a time that corresponds to half the round trip time of the
acoustic wave in Au.

To better understand the experimental results, we first have to explain how acoustic waves are generated in Au layers. The hot electron gas created due to the absorption of the pump pulses cools by rapidly heating the lattice. The rapid heating launches the acoustic wave. We know that the hot electron gas diffuses deeper into the Au layer due to the low electron-phonon coupling strength in gold. As a result, the spatial extent of the acoustic wave generated is much larger than the optical penetration depth (approximately 16 nm for 400 nm light) [83, 85]. For 100 nm and 200 nm thick Au layers, we expect the Au layer to more or less homogeneously heat up along the thickness of the layer. The resulting rapid expansion of the layer launches acoustic waves that causes an expansion and contraction of the lattice in a direction perpendicular to the surface.

The acoustic wave inside these Au layers can be seen as a standing wave formed by the addition of two counter-propagating acoustic waves, each having a wavelength corresponding to twice the thickness of the Au layer. In the 500 nm Au sample, the Au layer is not homogeneously heated due to the finite penetration depth of the electron gas energy. As a result, the spatial extent of the acoustic wave is approximately 300 nm. In all the measurements, the acoustic wave inside the Au layer remains grating-shaped because the lateral diffusion of heat can be ignored on the time scale of our experiments (< 1 ns) as the grating pitch is much larger than the thickness of the Au layer. Hence, in all of the samples, we have a grating-shaped acoustic wave bouncing back and forth in the Au layer.

The spatially periodic optical excitation of the Au layer gives rise to two effects: a thermal expansion of the layer and the launch of a grating-like acoustic wave. The thermal expansion leads to a quasi-static grating-shaped surface displacement, while the transient-grating acoustic echo results in a strongly time-dependent grating-shaped surface displacement. Both effects act as a phase grating as seen by the probe pulse. In the Au samples, the thermal expansion grating is formed about 10 ps after the optical excitation and is present for hundreds of picoseconds. In all of the measurements on Au samples, the displacement grating due to the acoustic echoes interferes with the thermal expansion grating present at the sur-
face. If only the thermal expansion grating gave rise to the diffraction of the probe pulse, then we would expect a quasi-constant diffraction signal from 10 ps to 700 ps. However, when the grating-shaped acoustic echo arrives at the air-Au interface, it destructively interferes with the quasi-constant thermal expansion grating. As a result, we observe a trough in the time-dependent diffraction signal. In short, in all of the measurements in Figure 5.1 (b), the troughs in the diffraction signal corresponds to the arrival of the acoustic echoes. The position of the first trough after 10 ps in all measurements matches the acoustic round trip time inside the Au layer. The best-fit model calculations for the measured data for all three Au layer thicknesses are shown by the black curves in Figure 5.1. The calculation is based on the model discussed in reference [18]. We obtain an excellent agreement between the measured and the calculated diffraction signal. It was observed in the calculation that the contribution from surface displacement (due to thermal expansion and acoustic wave) alone can explain the shape of the measured data very well. Note that the electron gas response is not included in the model but it is used as an input in the calculation. Therefore, only the data points for $t>10$ ps, when the electron gas has thermalized with the lattice, are used in the calculation.

To confirm that the arrival of acoustic echoes corresponds to the troughs in the diffraction signal, we performed experiments on the 500 nm thick Au sample, where we also measure the diffraction signal from the substrate side, as schematically shown in Figure 5.2 (a). Due to the finite penetration depth of the electron gas energy, heating of the lattice is expected to be absent near the substrate side, and the diffraction signal should be dominated only by the acoustic wave. The measured diffraction signals as a function of time are shown in Figure 5.2 (b). The red curve shows the diffraction signal when probed from the metal side (front probe), and the green curve shows the diffraction signal when probed from the substrate side (rear probe). The red curve in Figure 5.2 (b) is the same as the green curve in Figure 5.1 (b) but normalized to the peak at 145 ps. When measuring the diffraction signal from the glass side (green curve), we observe that the signal increases and reaches a maximum at $147 \pm 3$ ps, which coincides with the position of the peak in the red curve. Unlike the red curve, the green curve does not have a peak between 0-10 ps, associated with a grating in the electron
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Figure 5.2: (a) Schematic of the experiment. (b) Transient-grating pump-probe measurements on a 500 nm thick Au layer on a glass substrate. The red curve shows the diffraction signal when probed from the air-metal side (front probe) and the green curve shows the diffraction signal when probed from the substrate side (rear probe). The blue and black curves are the simulated diffracted signals.

temperature, showing that the electron gas energy does not diffuse to the rear of the sample.

When we probe from the glass side, we do not have any contribution to the diffraction signal from the thermal expansion grating. This is because the lattice temperature grating is very weak at the Au-glass interface. The hot electron gas energy created at the top surface of the Au layer does not diffuse deep enough into the 500 nm Au layer to give rise to a significant lattice temperature increase. Due to the absence of these effects, the peaks in the green curve correspond to the arrival of the acoustic wave at the Au-glass interface. As the troughs in the red curve correspond to the arrival of the acoustic wave at the air-metal side, the peak in the red curve is observed when the acoustic wave is farthest from the air-metal side, which is when the acoustic wave is near the Au-glass interface. Therefore, the peaks of the red and the green curve should coincide as we observe in the measurements.
The time separation between the peaks of the green curve is close to the acoustic round trip time inside 500 nm Au. The rise-time of the diffraction signal can be understood from the spatial extent of the acoustic wave generated inside the Au layer. As the rise-time of the first peak in the green curve is about 100 ps, we can estimate that the acoustic wave generated inside gold has a spatial extent of approximately 320 nm. These measurements supports our argument that the trough in the diffraction signal, when probed from the air-metal side, corresponds to the arrival of the acoustic echoes at the surface. This is also confirmed by the numerical simulation shown by the blue curve and the black curve for front and rear probing, respectively (Figure 5.2 (b)). These simulations also confirm that the main contribution to the diffraction signal is the grating-shaped displacement of the metal atoms. The diffraction of the 800 nm probe pulse due to the strain-optic and thermo-optic effect in Au is negligible. When probed from the glass side, we also expect a diffraction signal due to the grating-shaped acoustic wave propagating in the glass substrate. The grating-shaped acoustic wave can cause a change in the refractive index of the glass due to the strain-optic effect. Such a grating, propagating away from the metal-glass, can lead to oscillating diffraction signals as a function of time delay. This is commonly referred to as Brillouin scattering and is explained in detail in the next chapter. In this measurement shown in Figure 5.2 (green curve), we see hints of the oscillatory signal in the diffraction caused by Brillouin scattering in the glass. The effect is not very prominent because the diffraction due to the grating-shaped displacement of the interface dominates the total measured diffraction signal.

5.3.2 Acoustic waves in Ni and W

Now that we can launch and detect acoustic waves in Au layers, we performed experiments on metals which have higher electron-phonon coupling strengths than Au. As a result, we expect that the spatial extent of the generated acoustic wave to be smaller than that in Au. In this section, we show the results of transient-grating pump-probe measurements on flat nickel (Ni) and tungsten (W) layers on a glass substrate, when pumped and probed from the air-metal side (schematically shown in Figure 5.3 (a)). In Figure 5.3 (b), we plot the measured first-order diffraction signal
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Figure 5.3: (a) Schematic of the experiment. (b) Transient-grating pump-probe measurement on a 100 nm thick Ni layer. The red curve shows the measured diffracted signal and the blue curve shows the calculated diffracted signal.

as a function of the pump-probe delay for a 100 nm thick Ni layer on a glass substrate (red curve). We observe that the measured diffraction signal increases rapidly within 100 fs after optical excitation, and then drops to a lower value within 2 ps and drops further in 10 ps. The diffraction remains quasi-constant after 10 ps except for sharp periodic troughs at 29 ± 2 ps, 59 ± 2 ps, and 89 ± 2 ps. The rapid increase in diffraction signal around 0 ps and the decay within 2 ps, is due to the generation of a hot electron gas followed by the cooling of the electron gas by heating of the Ni lattice. The cooling of a hot electron gas in Ni is faster than for Au because the electron-phonon coupling strength in Ni is 12 times larger than that in Au [21].

Similar to the measurements on Au layers, in Ni the troughs in the diffraction signal correspond to the return of the longitudinal acoustic wave to the surface following reflection from the Ni-substrate interface. The multiple troughs in the diffraction signal show that the longitudinal acoustic wave bounces back and forth inside the Ni layer a few times. The position of the first trough (at 29 ps) and the time separation between the troughs (29-30
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ps) match the calculated acoustic round trip time in a 100 nm thick Ni layer, using an acoustic velocity of 5800 m/s [40]. The overall background diffraction signal seen between 2-120 ps, is due to the thermal expansion and the thermo-optic effect in Ni. The transient-grating pump pulses heat the Ni layer in a spatially periodic pattern, which results in a grating-like thermal expansion and a lattice temperature grating. This lattice temperature grating changes the refractive index of the Ni layer due to the thermo-optic effect. The delayed probe pulse can also diffract from this refractive index grating. The total diffracted signal is a coherent sum of the electric fields of the individual contributions, rather than a direct sum of diffraction efficiencies.

In Ni, the troughs in the diffraction signal caused by the acoustic echoes are sharper than in the case of Au. This can be explained by the stronger electron-phonon coupling strength and higher acoustic velocity in Ni. The hot electron gas generated within the optical penetration depth of 13 nm in Ni [122], rapidly cools to heat the lattice before the energy can diffuse deeper into the Ni. The generated acoustic wave is more localized in the propagation direction than in the case of Au. Hence the acoustic wave has a much higher frequency, and gives rise to sharper troughs in the diffraction signals. Our interpretation of the measured data is confirmed by the numerical simulation shown by the blue curve in Figure 5.3 (b), which is in good agreement with the measurements.

We also performed transient-grating pump-probe experiments on a 100 nm layer of tungsten (W) on glass, as schematically shown in Figure 5.4 (a). In Figure 5.4 (b), we plot the measured diffraction signal as a function of time delay for this sample (red curve). Similar to the measurement on the 100 nm Ni sample, we observe a sudden increase in the diffraction signal between 0-100 fs followed by a decrease within 2 ps. We see troughs in the diffraction signal at 32 ±2 ps and 64 ±2 ps. The troughs we observe are on top of a slowly decaying background diffraction signal. The troughs are caused by the return of the acoustic wave to the W surface after reflecting off the W-glass interface. The slowly decreasing background is due to the diffraction of the probe pulse from the grating-like thermal expansion of the layer and a lattice temperature grating (thermo-optic effect). The decrease in the background is due to the diffusion of lattice heat away from the
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The decay of the background is faster than in case of Ni most likely because W has higher thermal conductivity than Ni which leads to faster heat diffusion from the probing region. The fast decay of the electron dynamics signal between 0 ps and 2 ps, and the sharpness of the troughs at $32 \pm 2$ ps and $64 \pm 2$ ps, suggest that the acoustic wave in W has a high frequency. Note that the diffraction signal is normalized such that the maximum value is 1. The maximum diffraction signal is observed at $t=0$ ps due to the electron dynamics. However, this peak is not visible in this graph as the graph has been re-scaled such that the diffraction signals due to the acoustic echoes are visible.

Using the literature value for the acoustic velocity in poly-crystalline tungsten, $v=5.19$ nm/ps (=5190 m/s) [38], and measuring the time interval between the acoustic echoes (32 ps), we calculate the thickness of the W layer to be 84 nm, which is reasonably close to the thickness we aimed to deposit (100 nm). The discrepancy could be due to the error in the thickness measurement of the W layer during the physical vapor deposition. The numerical simulation also predicts the shape of the diffracted signal, and the shape of the measured diffraction signal due to the acoustic echoes in W.
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is consistent with the observations made by A. Devos and C. Lerounge [38].

5.3.3 Acoustic waves in Au-SiO$_x$-Au samples

Anticipating later measurements on more complex layer stacks, we gradually increased the complexity of our samples. In this section, we show the results from the transient-grating measurements on multi-layer samples with a layer of SiO$_x$ sandwiched between two Au layers, when pumped and probed from the air-Au side. In these experiments, we expect the SiO$_x$ layer to act as a barrier for the diffusion of the hot electron gas energy from the top layer to the bottom layer. This, in turn, would confine the initial lattice heating only to the top layer, and hence the acoustic wave will be generated only in the top Au layer. The acoustic wave then propagates through the SiO$_x$ to the bottom Au layer and returns to the top layer. The acoustic wave also undergoes partial reflection and transmission at the SiO$_x$-Au interface and Au-SiO$_x$ interface, which results in multiple acoustic echoes.

In Figure 5.5, we plot the measured diffraction signal from the two different samples. The first sample has a 60 nm Au layer on top of 7 nm SiO$_x$, on top of a 40 nm Au layer on a glass substrate (Figure 5.5 (a)). The second sample has a 60 nm Au layer on top of 7 nm SiO$_x$, on top of a 60 nm Au layer on a glass substrate (Figure 5.5 (b)). Both samples have the same thickness for the top Au layer. The measured diffracted signal of the first and the second sample are shown by the red and blue curve in Figure 5.5 (c). The red curve and blue curve have the same shape and strength until about 37 ps, after which they deviate. The rapid increase and the decrease of the diffraction signal during the first 10 ps is due to the electron dynamics in the 60 nm thick Au layer. The fact that both the curves have a very similar decay after the optical excitation means that hot electrons created in the top 60 nm layer of Au remain confined within that layer. The thin SiO$_x$ layer acts as a barrier for electron energy diffusion. The acoustic wave generated in both the samples will have the same frequency and strength because they are generated in the top Au layer, which in both samples is 60 nm thick. The diffraction due to grating-shaped thermal expansion of the Au layer will also be the same for both samples. The acoustic wave generated in the homogeneously heated 60 nm thick Au layer partially reflects from the
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Figure 5.5: (a) and (b) Schematics of the transient-grating pump-probe experiments on samples with 7 nm SiO\textsubscript{x} sandwiched between two Au layers. The top Au layer has the same thickness in both samples. (c) Measured diffracted signal as a function of pump-probe delay for the two samples.

Au-SiO\textsubscript{x} interface and returns to the air-Au interface. This results in the trough in the recorded diffraction signal at 37 ±1 ps. The large acoustic impedance mismatch between Au and SiO\textsubscript{x}, results in significant reflection of the acoustic wave at the interface. The acoustic impedance of Au and SiO\textsubscript{x} are 63.8 Ns/m\textsuperscript{3} and 13.5 Ns/m\textsuperscript{3}, respectively. The position of the first
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The acoustic wave partially transmitted at the Au-SiO\textsubscript{x} interface, propagates through the SiO\textsubscript{x} to the bottom Au layer, reflects from the Au-substrate interface and returns to the top surface. Since the thicknesses of the bottom layers are different, the shape of the measured diffraction signals after 37 ps is different. The difference is the shape of diffracted signals after 37 ps confirm that the acoustic wave has propagated beyond the SiO\textsubscript{x}, and the acoustic wave had reflected from the Au-glass interface. The shape of the signals is complicated due to acoustic waves bouncing back and forth in the top Au layer, bottom Au layer, and the whole stack.

Next, we performed transient-grating pump-probe measurements on two different samples with a thin layer of SiO\textsubscript{x} sandwiched between the Au layer. Here, the first sample has an 80 nm Au layer on top of 7 nm SiO\textsubscript{x}, which is on top of a 40 nm Au layer on a glass substrate (schematically shown in Figure 5.6 (a)). The second sample has a 60 nm Au layer on top of 7 nm SiO\textsubscript{x}, which is on top of a 60 nm Au layer on a glass substrate (schematically shown in Figure 5.6 (b)). In this case, the total thickness of the Au layer in both samples is the same. In Figure 5.6 (c), the red curve shows the diffraction signal from the first sample, and the blue curve shows the diffraction signal from the second sample.

The measured diffraction signal increases rapidly upon optical excitation and decays towards zero within the first 10 ps. We observe that the blue curve decays slower than the red curve. This observation is in agreement with the measurements shown in the previous chapter, where we observe a faster decay of the transient-grating signal for thicker Au layers. If the SiO\textsubscript{x} did not act as an electron energy barrier, then we would have observed a very similar decay of the signal for both samples, as both samples have the same total thickness of Au. The fact that we see a different decay rate when the top layer thicknesses are different, and the same decay rate when the top layer thicknesses are the same, implies that the 7 nm thick SiO\textsubscript{x} is an effective hot electron gas diffusion barrier.

The red and blue curves have different shapes after 10 ps. This difference arises from the fact that the arrival time of the first echo reflected from
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Figure 5.6: (a) and (b) Schematics of the transient-grating pump-probe experiments on samples with SiO$_x$ sandwiched between two Au layers. The total thickness of Au in both samples is the same. (c) Measured diffracted signal as a function of pump-probe delay for the two samples.

the SiO$_x$ layer is different. Also, the strength and spatial extent of the generated acoustic waves are different for both the samples. This can be explained by the difference in the thickness of the layer where the acoustic wave is generated. The two sets of measurements prove that we can launch an acoustic wave in a confined volume of Au, and the acoustic wave
can propagate through a multi-layer sample containing a dielectric layer. Complete numerical simulation of the diffraction signal for these Au-SiO$_x$-Au samples were not performed as the primary goal of these experiments were to understand electron energy diffusion and to check whether acoustic waves can propagate across a metal-dielectric interface.

5.4 Conclusion

We have shown that the transient-grating pump-probe setup can be used to launch and detect acoustic waves in flat Au, Ni, and W layers. We observe a background thermal grating, either due to thermal expansion of the layer or thermo-optic effect, on which the generated acoustic wave becomes visible as troughs when they arrive at the surface. Further support for this is found in experiments, where we pump from the air-metal side but probe from the substrate side. We also show that Ni can be used as an acoustic transducer to launch high-frequency acoustic waves in Au. Our experiments on samples that have a 7 nm thick layer of SiO$_x$ sandwiched between two Au layers demonstrate that a thin layer SiO$_x$ can be an effective electron energy diffusion barrier.