Detection of hidden gratings using light and sound

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3 EXPERIMENTAL DETAILS

3.1 Laser system

Most of the pump-probe measurements discussed in this thesis were performed using a Ti:Sapphire amplifier (Femtopower, Spectra-Physics). It is a 1 kHz-repetition rate, multi-pass amplifier seeded with broadband, ultra-short pulses of about 10 fs pulse duration at a repetition rate of approximately 79 MHz from a dispersion controlled oscillator. A functional block diagram of the laser system is shown in Figure 3.1. The pulses from the oscillator are stretched by propagation through thick layers of glass prior to amplification to avoid damaging the Ti:Sapphire crystal inside the amplifier. The pulses are then amplified by multiple passes through the crystal, pumped by a 22 W diode-pumped Nd:YLF laser operating at a wavelength of 527 nm and a repetition rate of 1 kHz. After the four passes, a single pulse is selected from the MHz-pulse train with two Pockel’s cells. This pulse is sent through Fastlite’s DAZZLER, an acousto-optic programmable filter that is used to pre-compensate third- and fourth-order dispersion (i.e. optimize the re-compressed pulse-width). The DAZZLER also spectrally shapes the optical pulse in order to reduce the effect of gain-narrowing in the amplifier. Further amplification of this pulse to 4 mJ is achieved by
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Figure 3.1: Functional block diagram of the laser system.
six further passes through the Ti:Sapphire crystal. After the 10th pass, the pulse is picked off and sent to a grating compressor followed by a set of chirped mirrors for pulse compression. Finally, the average output power from the amplifier is 3 W at 1 kHz repetition rate, and pulses have a duration of 30 fs with a wavelength centered around 800 nm.

3.2 Pump-probe setup

Figure 3.2: Two configurations of pump-probe setup: (a) single pump pulse excites the sample, and the probe pulse diffracted from the ”copy” of the buried grating is measured as a function of pump-probe delay. (b) two pump pulses interfere to form a grating, and the diffracted probe pulse is measured as a function of time.

All of the experiments discussed in the thesis were performed using various configurations of a pump-probe setup. Pump-probe setups are usually employed to study extremely fast processes occurring in the physical, chemical, and biological research fields. In our pump-probe setups, a beamsplitter splits a laser pulse into a ‘pump’ pulse and a ‘probe’ pulse. Both of the two pulses arrive at the sample via different paths, and the optical path difference between the pulses is varied to create a temporal delay between the pump and the probe. This is done by reflecting light off two mirrors in a retro-reflecting geometry mounted on a mechanical delay line. The pump pulse excites the sample and can change the physical properties of the sample, while the delayed probe pulse detects these changes. In our experiments, we mainly use two schemes of a pump-probe setup. In the first scheme shown in Figure 3.2 (a), a single pump pulse excites a flat metal layer with an optically buried grating. The shape of the buried grating is transferred to the surface of the metal layer in the form of a spatially
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periodic surface displacement or a spatially periodic change of the complex refractive index. The delayed probe pulse diffracts from this “copy” of the buried grating, and the first-order diffracted signal is recorded. Various physical mechanisms responsible for creating the “copy” of the buried grating are discussed in Chapter 3, 6 and 7. In the second scheme shown in Figure 3.2 (b), two pump pulses overlap spatially and temporally to form an interference pattern that excites the sample in a grating-like pattern. The probe pulse diffracts due to the change in the optical constants of the sample or by a spatially periodic displacement of the surface induced by the periodic excitation (details can be found in Chapter 4 and 5). In laser-induced ultrasonics experiments, the diffraction can also be due to the spatial periodic displacement of the surface. In both the schemes, we measured the intensity of the diffracted probe pulse with a silicon photodetector. The photodetector generates a voltage proportional to the intensity of the light recorded by the photodiode. Fluctuations in the output energy of the laser pulses are accounted for by dividing the measured diffracted signal by a reference signal obtained by a silicon photodetector that monitors pulse-to-pulse variations in the output energy. The signals are sent to a boxcar integrator, which integrates the electronic signals. The analog signals from the boxcar integrator are converted to a digital signals using an analog-to-digital converter. The data acquisition card triggered by the laser pulses reads out the voltages and also records the position of the mechanical delay line. The home-built software can vary the pump-probe delay in small steps (down to 1 fs) and simultaneously records the reference signal and the diffracted signal. The software can average the signal over a number of optical pulses for a given time delay.

3.3 Determining the pump-probe temporal overlap

3.3.1 Sum-frequency generation

Before using the transient-grating pump-probe setup for experiments, we have to determine the exact position of the mechanical delay stage, for which the pump and probe pulses overlap temporally on the sample. For this purpose, a type 1 beta barium borate (BBO) crystal was placed at the
3.3. Determining the pump-probe temporal overlap

Figure 3.3: The crystal orientation and polarization of the 400 nm and 800 nm wavelength beams used to generate 266 nm wavelength beam.

location of the sample where we want the pump and probe pulses to overlap. When the 400 nm wavelength pump pulse and the 800 nm wavelength probe pulse overlap both spatially and temporally inside the BBO, we expect 266 nm light to be created as a result of sum-frequency generation inside the BBO crystal. The BBO used here was 200 µm thick and 6 mm × 6 mm in area and the crystal orientation was $\theta = 44.3^0$ and $\phi = 90^0$ so that, the crystal can generate a 266 nm wavelength beam when a 400 nm and 800 nm wavelength beams are incident perpendicular to the surface of the BBO crystal. The crystal orientation and polarization of the 400 nm and 800 nm wavelength beams with respect to the BBO crystal, are shown in Figure 3.3. We focus the 400 nm wavelength pump beam and the 800 nm wavelength probe beam into the BBO in a non-collinear geometry such that both beams overlap spatially inside the crystal. The angle between the beams was $2.5^0$. The 400 nm and 800 nm wavelength light and the generated 266 nm wavelength light is transmitted through the crystal. We placed a SiC detector that measures the intensity of the 266 nm beam at the location where we expect the 266 nm beam. Spatially, the 266 nm wavelength beam is expected to be generated in between the 400 nm and
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800 nm wavelength beams but not exactly in the middle. In the detection plane, if the distance between the 400 nm beam and the 800 nm beam is x, then the 266 nm wavelength beam will be positioned at roughly x/3 from the 400 nm beam. This is a direct consequence of the momentum conservation between the 800 nm, 400 nm and 266 nm photons.

To determine the temporal overlap, the delay between the 400 nm pump pulse and the 800 nm probe pulse is varied while observing the presence and intensity variation of the generated 266 nm light. When the pulses are not overlapping, we don’t expect any 266 nm wavelength light to be generated. As the pump and probe pulse begin to overlap in time, the intensity of the 266 nm wavelength light increases. The pump-probe delay time for which the highest intensity is recorded is when the pump and probe perfectly overlap in time. Although the 266 nm wavelength is invisible to the human eye, we can detect the generation of this UV light by placing a white paper where we expect the beam. When the 266 nm wavelength light falls on a white paper, we can see a violet-colored spot due to the fluorescence of the white paper. For experiments that use two pump pulses, the same procedure described above is repeated with the second 400 nm pulse and the 800 nm pulse. The delay between the two 400 nm pulses is adjusted until all three pulses overlap spatially and temporally.

3.3.2 Transient grating in BBO

Now that we know the spatial and temporal position where the two pump pulses and the probe pulse overlaps, we can perform transient-grating pump-probe experiments. To align the location of the detector such that it measures the first-order diffracted probe pulse, we performed the first set of transient-grating experiments on the BBO crystal that was used to find the temporal overlap of the pulses. When two pump pulses overlap on the BBO crystal, a spatially periodic intensity pattern with a grating pitch of 6 µm is created. The intensity of the 400 nm wavelength pump pulses was increased in order to induce spatially periodic changes in the refractive index of the BBO, which arise from nonlinear optical effects in the crystal. The 800 nm wavelength probe pulse then diffracts from the refractive index grating, and the first-order diffracted beam can be recorded by the detector. This experiment can be performed either in transmission or in
3.4 Sample preparation

The metal layers of the samples used in the measurements were fabricated by various physical vapor deposition (PVD) techniques, such as thermal evaporation, electron beam evaporation, and sputter deposition. All of the samples were fabricated at the Amsterdam Nanocenter at AMOLF.

The Au layers were mostly deposited using a thermal evaporator called “Nanoontje”. The evaporator uses a resistive heat source to evaporate the metal in a vacuum environment. The evaporated metal forms a thin film

Figure 3.4: Diffracted signal from the BBO crystal as a function of pump-probe delay.
3. Experimental details

on the substrate placed above the heat source. The Au and Cu layers in the samples with buried gratings, discussed in Chapter 4, were fabricated using the same evaporator. The SiO$_x$ layer in samples discussed in Chapter 5 were also fabricated using this evaporator.

The W and Ni layers on the samples discussed in Chapter 5 were deposited using an electron beam evaporator called “Kameleon”. Instead of using a resistive heat source, in electron beam deposition, the target metal is bombarded with an electron beam from a charged tungsten filament to melt and evaporate the metal. The evaporated metal forms a thin film on the substrate placed above the metal target. This evaporation technique is better suited for metals having a high melting point. The bilayer metal samples discussed in Chapter 4, which contain both Au and Pt layers, were deposited using this evaporator. The Ni layers in samples discussed in Chapter 7 were deposited using another electron beam evaporator called “E-flex”.

The SiO$_2$ and Si$_3$N$_4$ layers in the samples discussed in Chapter 6 were deposited using reactive sputter deposition. In sputter deposition, the material ‘target’ is subjected to an argon (Ar) gas plasma in a relatively low vacuum ($10^{-2}$ mbar). The energetic Ar ions from the Ar plasma bombard the target material and remove (sputter) atoms from the material. The removed atoms are then deposited onto the substrate to form a uniform thin film. In reactive sputtering deposition, the ejected material undergoes a chemical reaction before reaching the substrate. This is done by introducing a reactive gas in the deposition chamber. For the deposition of SiO$_2$, a Si target is sputtered in the presence of oxygen gas (O$_2$) and for deposition of Si$_3$N$_4$, a Si target is sputtered in the presence of nitrogen gas (N$_2$). The composition of the material can be adjusted by varying the ratio between the Ar gas and the reactive gas (O$_2$ or N$_2$). The parameters used during the deposition of 18 nm SiO$_2$ and 18 nm Si$_3$N$_4$ are shown in Table 3.1.

The gratings on the metal layer of the samples discussed in Chapter 6 and 7 were fabricated by UV lithography. The resist, S1805, was spin-coated on the metal layer for approximately 45 seconds at 2000 rpm. The resist-coated sample was then exposed with UV light through the mask containing the grating pattern for 2-3 seconds using a commercial UV mask aligner,
### 3.4. Sample preparation

<table>
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<th>SiO$_2$</th>
<th>Si$_3$N$_4$</th>
</tr>
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<tr>
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<td>$1 \times 10^{-2}$</td>
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<td>Nitrogen flow rate (sccm)</td>
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<tr>
<td>RF Power (W)</td>
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<td>143</td>
</tr>
<tr>
<td>Time (s)</td>
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<td>1700</td>
</tr>
</tbody>
</table>

Table 3.1: Parameters for the deposition of 18 nm SiO$_2$ and Si$_3$N$_4$ using reactive sputter deposition.

Figure 3.5: Optical microscope image of a 6 µm pitch, 50% duty cycle grating fabricated on a flat gold layer.

“Suss MA6”. The exposure wavelength is 385 nm, and the system operated at 25 mW of optical power. The resist is then developed in the developer solution, Ma-D 533-S, for approximately 30 seconds. Another layer of metal is deposited on top of the patterned resist layer, and the remaining resist layer is removed by immersing the whole sample in a “lift-off” solution. The optical microscope image of a grating on a Au layer fabricated with this recipe is shown in Figure 3.5. The grating shown here has a pitch of 6 µm, and a duty cycle of approximately 50%.