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Understanding and eliminating artifact signals from diffusely scattered pump beam in measurements of rough samples by time-domain thermoreflectance (TDTR)

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Time-domain thermoreflectance (TDTR) is a pump-probe technique frequently applied to measure the thermal transport properties of bulk materials, nanostructures, and interfaces. One of the limitations of TDTR is that it can only be employed to samples with a fairly smooth surface. For rough samples, artifact signals are collected when the pump beam in TDTR measurements is diffusely scattered by the rough surface into the photodetector, rendering the TDTR measurements invalid. In this paper, we systemically studied the factors affecting the artifact signals due to the pump beam leaked into the photodetector and thus established the origin of the artifact signals. We find that signals from the leaked pump beam are modulated by the probe beam due to the phase rotation induced in the photodetector by the illumination of the probe beam. As a result of the modulation, artifact signals due to the leaked pump beam are registered in TDTR measurements as the out-of-phase signals. We then developed a simple approach to eliminate the artifact signals due to the leaked pump beam. We verify our leak-pump correction approach by measuring the thermal conductivity of a rough InN sample, when the signals from the leaked pump beam are significant. We also discuss the advantages of our new method over the two-tint approach and its limitations. Our new approach enables measurements of the thermal conductivity of rough samples using TDTR. Published by AIP Publishing.

I. INTRODUCTION

Time-domain thermoreflectance (TDTR)\(^1\)\(^--\)\(^3\) is a robust pump-probe technique that is widely applied to measure the thermal conductivity of bulk materials\(^4\)\(^--\)\(^6\) and nanostructures\(^7\)\(^--\)\(^9\) and the thermal conductance of interfaces.\(^10\)\(^--\)\(^14\)

A schematic diagram of the TDTR setup in our lab is shown in Fig. 1. In a typical TDTR setup, laser pulses from a Ti:sapphire laser are split into pump and probe beams. The pump beam, modulated at a modulation frequency \(f\) by an electro-optical modulator (EOM), heats the sample periodically. The temperature response at the sample surface at frequency \(f\) is then monitored by the time-delayed probe beam measured by a Si photodiode detector, via change of reflectance with temperature (i.e., thermoreflectance, \(dR/dT\)). The value of \(dR/dT\) of the transducer metals used in TDTR measurements is, however, usually small, on the order of \(10^{-4}\)\(--\)\(10^{-5}\) \(\text{K}^{-1}\).\(^15\) To measure this small change of intensity of the reflected probe beam, a radio-frequency (RF) lock-in amplifier is employed to pick up signals only at \(f\) and reject signals and noises at other frequencies. However, the RF lock-in amplifier does not reject signals from the pump beam that is also modulated at \(f\). Thus, in typical TDTR measurements, special care is given to ensure that the photodetector does not collect any reflected light from the pump beam. For smooth samples, this can be conveniently achieved by blocking the specularly reflected pump beam using an aperture. For rough samples, however, the pump beam is diffusely scattered and thus cannot be blocked by an aperture. Due to the small value of \(dR/dT\) (\(\sim 10^{-4}\) \(\text{K}^{-1}\)), the artifact signals, due to the diffusely scattered pump beam that is collected by the photodetector (often called the leaked pump beam), could strongly affect TDTR measurements even if its intensity is \(<0.01\%\) than that of the reflected probe beam.

Usually, the artifacts due to the leaked pump beam are eliminated via either a two-tint\(^16\) or a two-color approach.\(^17\) In the two-color approach, either the pump or the probe beam is frequency-doubled through second harmonic generation. The leaked pump beam can then be easily rejected using an appropriate optical filter. In the two-tint approach, on the other hand, the pump and probe beams with slightly different wavelengths are created using sharp-edged bandpass and lowpass optical filters.\(^16\) The diffusely scattered pump beam is rejected by placing another sharp-edged filter before the photodetector. Due to less complex instrumentation of the two-tint approach and smaller laser spot sizes that can be achieved, we usually use the two-tint approach to measure rough samples in our lab. One problem for the two-tint approach is that due to sharp edges of the optical filters, fluctuations in the wavelength of the laser often translate into fluctuations in the intensity of the pump and probe beams. Thus, the signal-to-noise ratio (SNR) of two-tint measurements could be significantly reduced from what could be achieved without the

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sharp-edged filters, especially when we need to increase the spectral separation of the pump and probe beams to reject a large amount of leaked pump beams as we measure very rough samples.

While the artifact signals due to the leaked pump beam are ubiquitously observed in different labs, the reasons why the artifact signals are even captured during TDTR measurements are still unknown. In a double-modulation TDTR setup (as in our lab), the pump beam is modulated at a radio frequency (e.g., 10 MHz), while the probe beam is modulated at an audio frequency (AF) (e.g., 200 Hz). So, in addition to the RF lock-in amplifier, a second audio frequency (AF) lock-in amplifier is used to eliminate any artifact signals (e.g., coherent pick-ups by the cables) captured by the RF lock-in amplifier. Since the pump beam does not have any audio-frequency components, any artifact signals from the leaked pump beam should be rejected by the second AF lock-in amplifier. Also, it is interesting that the artifact signals due to the leaked pump beam are only observed when the probe beam is not blocked. This shows that the leaked pump beam is modulated at the audio frequency by the probe beam, but it is still unclear at which point in TDTR measurements that the modulation occurs.

In this work, we systemically studied the factors affecting the artifact signals in TDTR measurements due to the leaked pump beam and successfully identified the source of modulation of the leaked pump beam at the audio-frequency of 200 Hz. We find that, when we intentionally increase the leaked pump beam, the in-phase TDTR signal $V_{\text{in}}$ is essentially unchanged while the absolute value of out-of-phase TDTR signal $V_{\text{out}}$ increases significantly. To better understand the observation, we define a proportionality constant $\alpha = -\Delta V_{\text{out}}/\Delta V_{\text{leak}}$ to quantify the change of $V_{\text{out}}$ in TDTR measurements with the amplitude of the leaked pump beam registered by the RF lock-in amplifier $V_{\text{leak}}$. We find that $\alpha$ only depends on the photocurrent of the photodiode and the Q-factor of the resonant circuit in the TDTR setup. We then experimentally demonstrate that such dependence is due to a slight phase rotation in the phase of the leaked pump beam induced by the reflected probe beam. This phase rotation provides the necessary modulation to the leaked pump beam at the audio frequency of the AF lock-in amplifier. We then developed a simple correction approach to eliminate the artifact signals of the leaked pump beam and thus enable measurements of the thermal conductivity of rough samples using TDTR. We apply this method on a rough InN sample and successfully obtain the correct thermal conductivity for the sample. We show that the pump-leak correction approach could be applied to almost all homogeneously rough samples. We also demonstrate that, for the same level of leaked pump beam, our approach yields much higher signal-to-noise ratio compared to that of the two-tint method and thus establishes the superiority of the new approach.

II. ARTIFACT SIGNALS FROM LEAKED PUMP BEAM

Fig. 1 shows a schematic diagram of the TDTR setup in our lab. Here, we focus on discussing the signal processing in TDTR and readers are referred to Ref. 18 for detailed descriptions on how we acquire and analyze TDTR measurements. Our laser is a femtosecond Ti:sapphire ultrafast laser with a repetition rate of $1/\tau = 80$ MHz. In the frequency domain, laser pulses from the ultrafast laser are represented by a series of delta functions at multiples of $1/\tau$. We employ a double-modulation approach in our TDTR measurements. In the double-modulation scheme, the pump beam is modulated by an EOM at a radio frequency $f_a$ (e.g., 10 MHz), while the probe beam is modulated by a mechanical chopper at an audio frequency of $f_a = 200$ Hz. Due to the RF modulation, both the pump beam and the temperature oscillation at the sample surface induced by the pump beam have frequency components of $m/\tau \pm f_a$, where $m$ is an integer. (Here, we do not consider components at higher harmonics of $f_a$, since they are removed by a RLC resonant circuit, see below.) On the other hand, since the temperature oscillation is monitored by the probe beam via thermoreflectance, the reflected probe beam, measured by a photodiode, is modulated at both the radio frequency $f_a$ and the audio frequency $f_a$. Thus, the reflected probe beam has frequency components of $m/\tau \pm f_a \pm f_a$. (Again, we do not consider components at higher harmonics of $f_a$, since they are not picked up by the AF lock-in amplifier, see below.) We use a 30 MHz lowpass filter to remove signals at higher multiples of $1/\tau$ and use a RLC resonant circuit to eliminate the higher harmonics of $f_a$. (The RLC resonant circuit is formed by serially connecting an inductor to the photodiode, with the photodiode acting as a capacitor.) The RLC resonant circuit also enhances signals around $f_a$ by a factor of Q, the quality factor of the resonant circuit.
Even after the amplification by the RLC resonant circuit and a pre-amp, the signals due to the temperature oscillation are still comparably smaller than the signals at $1/\tau$ and the noise due to the small $dR/dT$ ($10^{-2}$ to $10^{-3}$ K$^{-1}$) of the metal transducer. Thus, we employ a RF lock-in amplifier to pick up only the minute signals due to the temperature oscillation at sample surface at $\pm f_r \pm f_a$. In the RF lock-in amplifier, we set the time constant to 100 μs, so that the bandwidth of the RF lock-in amplifier ($\sim$ 3 kHz) is sufficiently large to capture the sidebands due to modulation at the audio frequency $f_a$. However, at this point, artifact signals at $f_r$ will also be picked up by the RF lock-in amplifier. The sources of the artifact signals include, for example, coherent pickups due to RF electromagnetic waves in resonance with the cables and electronic devices in the TDTR setup, and a small amount of pump beam leaked into the photodetector. After demodulation at the radio frequency $f_r$, the in-phase and out-of-phase outputs of the RF lock-in amplifier, now only with frequency components of $\pm f_a$, are fed into an AF lock-in amplifier. Since the AF lock-in amplifier only pick up the signals with an $f_a$ component, the artifact signals should be rejected by the AF lock-in amplifier as they lack $\pm f_a$ components.

We find that even with the double-modulation approach, the artifact signals from the pump beam leaked into the photodetector are still registered by the AF lock-in amplifier in TDTR measurements. To demonstrate this point, we monitored the in-phase $V_{in}$ and the out-of-phase $V_{out}$ TDTR signals of a 99.99% pure copper coated with a 91-nm-thick Al film, while the pump beam was intentionally and systematically leaked into the photodetector by adjusting the position of the aperture, see Fig. 1. We plot $V_{in}$ and $V_{out}$ registered by the AF lock-in amplifier, as a function of the signal due to the leaked pump beam $V_{leak}$, registered by the RF lock-in amplifier, in Fig. 2. We can directly compare $V_{in}$, $V_{out}$ and $V_{leak}$, since they all are amplified by the same factor after being measured by the photodetector. Note that while $V_{in}$ and $V_{out}$ are TDTR signals registered by the AF lock-in amplifier, $V_{leak}$ is not a TDTR signal but is essentially an intensity of the leaked pump beam measured by the photodetector. We set the phase in the reference channel of the RF lock-in amplifier when $V_{leak}$ = 0 and kept the same phase as we increased $V_{leak}$. We find that as $V_{leak}$ increases, the magnitude of $V_{out}$ increases linearly, see Fig. 2. Interestingly, we find that even when $V_{leak}$ is >10 times larger than $V_{in}$, $V_{in}$ is still independent of $V_{leak}$. This trend is not confined to the measurements of the Al/Cu sample that we present in Fig. 2 but generic to all samples we measured. The change of $V_{out}$ due to $V_{leak}$ suggests that the leaked pump beam is modulated at $f_a$ and thus registered by the AF lock-in amplifier.

We postulate that modulation of the leaked pump beam at $f_a$ could occur at either sample surfaces or at the photodetector, since both the pump and the probe beams cross paths at these locations. If the modulation of the pump beam occurs at sample surfaces, properties of the samples (i.e., substrates and the metal transducer) may affect the artifact signals due to the leaked pump beam. On the other hand, if the modulation occurs at the photodetector, properties related to the photodetector such as photocurrent of photodetector and Q-factor of the RLC resonant circuit may affect the artifact signals.

To better pinpoint the origins of the artifact signals of the leaked pump beam, we define a proportionality constant $\alpha = -\Delta V_{out}/\Delta V_{leak}$ to quantify the change of the out-of-phase signal ($V_{out}$) due to the leaked pump beam. We measured $\alpha$ for 3 samples (Al/Cu, Al/SiO$_2$, and Cu/SiO$_2$, where the transducer films are ~90 nm thick) using a wide range of experimental parameters mentioned above. We achieve different photocurrents by changing the intensity of the reflected probe beam and the responsivity of the photodetector (by changing the wavelength of the probe laser). We estimate the photocurrent of the photodetector by measuring the voltage across a 50 Ω resistor serially connected to the photodetector. We modify the Q-factor and the resonant frequency $f_r$ of the RLC circuit by changing the resistor and the inductor in the RLC circuit. We determine the Q-factor of the RLC resonant circuit from the full-width-half-maximum of the resonance by measuring the TDTR signals at frequencies around $f_r$ using the RF lock-in amplifier. In addition, we also removed the RLC resonant circuit and measured $\alpha$ at two modulation frequencies $f_r$ (i.e., 1 MHz and 10 MHz, see the solid symbols in Fig. 3(b)) without the resonant circuit.

We summarize the measured $\alpha$ in Fig. 3, as a function of the photocurrent and Q-factor. We find that the proportionality constant $\alpha$ from the leaked pump beam does not depend on the type of samples and transducers, see Fig. 3(a), suggesting that the modulation of the leaked pump beam at $f_a$ does not occur at the sample surfaces. On the other hand, we find that $\alpha$ depends strongly on the properties related to the photodetector, i.e., photocurrents in the photodetector and Q-factor of the RLC circuits. The dependences on both the photocurrent and the Q-factor are linear, see Fig. 3. Since both factors are affected by the photodetector, Fig. 3 suggests that the modulation of the signals from the leaked pump beam at $f_a$ occurs at the photodetector. Interestingly, we also find that $\alpha$ is small but nonzero after we remove the RLC resonant circuit, see Fig. 3(a).
One possible explanation to the observed modulation of the leaked pump beam at the photodetector is that the probe beam prompts a phase rotation in the \( \Delta \theta \) phase of the leaked pump beam in the RF lock-in amplifier. As the pump and probe beams do not overlap, TDTR signals (i.e., \( V_{\text{in}} \) and \( V_{\text{out}} \)) are zero. Thus, we can directly measure \( \Delta V_{\text{in, leak}}^{\text{in}} \) and \( \Delta V_{\text{out, leak}}^{\text{out}} \) from the outputs of AF lock-in amplifier. As expected, \( \Delta V_{\text{leak}}^{\text{in}} \approx 0 \) as suggested by Eq. (1). We then calculate \( \Delta \theta \) from \( \Delta V_{\text{leak}}^{\text{out}} \) and \( \Delta V_{\text{in}} \) using Eq. (1). In the second approach, we directly measured the phases of the leaked pump beam in the RF lock-in amplifier, with and without illumination of the probe beam, and derived \( \Delta \theta \) from the difference in the measured phases. The phase of the leaked pump beam is readily determined from the phase of the reference channel of the RF lock-in amplifier when the out-of-phase channel is zero. The uncertainty of \( \Delta \theta \) is estimated from the standard deviation of the outputs of the RF lock-in amplifier. We compare \( \Delta \theta \) derived using both approaches to \( \alpha \) measured at the same photocurrent and Q in Fig. 4. We note that \( \Delta \theta \) derived using the second approach is \( \sim 10\% \) higher than \( \Delta \theta \) derived using the first approach, see Fig. 4. This small difference could be due to, e.g., imperfect modulation of the leaked pump beam as a result of a finite rise time of the photodetector. We confirm that \( \Delta \theta = \alpha \) within experimental uncertainty for \( \Delta \theta \) derived using both approaches, see Fig. 4.

We are not certain of the sources of the phase rotation. One possible reason is that the junction capacitance of the p-i-n photodiode is slightly changed while illuminating and thus alters the resonance of the RLC circuit. However, this cannot explain why we still observe the phase rotation even without the RLC resonant circuit.

III. TDTR MEASUREMENTS OF ROUGH SAMPLES

We can easily correct the artifact signals due to leaked pump beam using Eq. (1) and thus enable TDTR measurements. We vary a controlled amount of pump beam leaked into the photodetector. As the pump and probe beams at sample surfaces by \( \sim 25 \mu m \) and perform TDTR measurements with a controlled amount of pump beam leaked into the photodetector. As the pump and probe beams do not overlap, TDTR signals (i.e., \( V_{\text{in}} \) and \( V_{\text{out}} \)) are zero. Thus, we can directly measure \( \Delta V_{\text{in, leak}}^{\text{in}} \) and \( \Delta V_{\text{out, leak}}^{\text{out}} \) from the outputs of AF lock-in amplifier. As expected, \( \Delta V_{\text{leak}}^{\text{in}} \approx 0 \) as suggested by Eq. (1). We then calculate \( \Delta \theta \) from \( \Delta V_{\text{leak}}^{\text{out}} \) and \( \Delta V_{\text{in}} \) using Eq. (1). In the second approach, we directly measured the phases of the leaked pump beam in the RF lock-in amplifier, with and without illumination of the probe beam, and derived \( \Delta \theta \) from the difference in the measured phases. The phase of the leaked pump beam is readily determined from the phase of the reference channel of the RF lock-in amplifier when the out-of-phase channel is zero. The uncertainty of \( \Delta \theta \) is estimated from the standard deviation of the outputs of the RF lock-in amplifier. We compare \( \Delta \theta \) derived using both approaches to \( \alpha \) measured at the same photocurrent and Q in Fig. 4. We note that \( \Delta \theta \) derived using the second approach is \( \sim 10\% \) higher than \( \Delta \theta \) derived using the first approach, see Fig. 4. This small difference could be due to, e.g., imperfect modulation of the leaked pump beam as a result of a finite rise time of the photodetector. We confirm that \( \Delta \theta = \alpha \) within experimental uncertainty for \( \Delta \theta \) derived using both approaches, see Fig. 4.

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measurements on rough samples. To achieve the pump-leak correction, we simultaneously monitor the intensity of the leaked pump beam $V_{\text{leak}}$ registered by the RF lock-in amplifier, while we perform TDTR measurements. In our setup, this can be readily achieved by monitoring the offsets of the square wave inputs to the AF lock-in amplifier, in addition to the amplitudes of the square waves usually monitored during typical TDTR measurements; $V_{\text{leak}}$ equals the offset of the input to the in-phase channel. To ensure a sufficiently high signal-to-noise (>50) for the $V_{\text{leak}}$ measured by this approach, we set the time constant of the AF lock-in amplifier to 700 ms.

In conventional TDTR, the phase of the reference channel of RF lock-in amplifier is determined by the fact that the out-of-phase signal $V_{\text{out}}$ is constant when the delay time crosses $t = 0$. When there is a leaked pump beam, $V_{\text{out}}$ is composed of the “real” out-of-phase TDTR signal and artificial signal $\Delta V_{\text{out, leak}}$ from the leaked pump beam. As we mentioned in Sec. II, $\Delta V_{\text{out, leak}}$ is independent of delay time. Thus, we can still use the same way as conventional TDTR to determine the phase when there is a leaked pump beam and applying our pump-leak correction approach to measure rough samples.

We test our pump-leak correction approach on an Al/InN/GaN/sapphire sample. The InN film is 2.5 μm thick and was deposited by a molecular beam epitaxy (MBE), provided to us by Prof. Gregor Koblmueller of Technical University of Munich (TUM). The sample is rough because a high lattice constant mismatch between InN and the substrate produces a high concentration of line defects in the InN film and a rough surface morphology. We coated the InN sample with ~98 nm thick Al by thermal evaporation. The thickness of Al is determined by picosecond acoustics as in conventional TDTR, since the in-phase signal $V_{\text{in}}$ is hardly affected by the leaked pump beam. To validate our new approach, we performed conventional TDTR, two-tint TDTR, and TDTR with the pump-leak correction on the rough InN sample. In all the TDTR measurements, we used laser 1/e² radii of 4.5 μm, a modulation frequency $f_r$ of 10 MHz for the pump beam, and an audio-frequency $f_a$ of 200 Hz for the probe beam. We employed a total laser power of 60 mW to ensure that the temperature rise on the sample is <5 K. We used appropriate natural density filters before the photodetector to fix the photocurrent of the photodetector at ~0.4 mA.

We performed TDTR measurements at two spots (Spots A and B) on the rough InN sample. Spot A (Fig. 5(a)) is relatively smooth with 96% of light incident on the spot being specularly

![FIG. 5. Bright-field microscope images of an Al/InN/GaN/sapphire sample when TDTR measurements were performed at (a) relatively smooth Spot A and (b) rough Spot B. The white dots in the images are the laser spots. (c) $V_{\text{in}}, V_{\text{out}},$ and $V_{\text{leak}}$ measured at Spot B (black), as labeled. $V_{\text{out}}$ is instantaneously corrected using $\Delta V_{\text{out, leak}}$ calculated from Eq. (1) (red) in our pump-leak correction approach. (d) Conventional TDTR measurements at Spot A (olive diamonds, labeled “Spot A”) and Spot B (black circles, labeled “Spot B”), compared to measurements at Spot B using the pump-leak correction approach (red circles, labeled “pump-leak correction”) and the two-tint approach (blue triangles, labeled “two-tint”). Solid lines are calculations using a thermal model assuming $\Lambda = 98$ W m⁻¹ K⁻¹ (red) and $\Lambda = 62$ W m⁻¹ K⁻¹ (black), respectively, while the dashed lines are calculations with the $\Lambda$ varied by ±10%. For all the measurements, we use a modulation frequency of 10 MHz ($Q = 11$), a spot size of 4.5 μm, a total power of 60 mW, and a photocurrent of 0.4 mA.](image-url)
reflected. We find that $V_{\text{leak}} < 0.3V_{\text{out}}$ at this spot, implying that $\Delta V_{\text{out}}$ is less than 1% of $V_{\text{out}}$. We derive the thermal conductivity of InN, $\Lambda = 98$ W m$^{-1}$ K$^{-1}$, from conventional TDTR measurements at Spot A, see the olive diamonds in Fig. 5(d). The result is in good agreement with our previous measurement on a 1-μm-thick InN film and is ~20% smaller than the measured thermal conductivity of 0.5-2.1 μm thick InN films. Spot B (Fig. 5(b)), on the other hand, is rough, with only 80% of light incident on the spot being specularly reflected. At this spot, $V_{\text{leak}} > 6V_{\text{out}}$, see Fig. 5(c). Due to the high level of leaked pump beam, conventional TDTR measurement yields a wrong result of only 62 W m$^{-1}$ K$^{-1}$, see the black circles in Fig. 5(d). We thus applied the pump-leak correction approach that we developed to derive the thermal conductivity at this spot. We simultaneously measured $V_{\text{leak}}$, $V_{\text{in}}$, and $V_{\text{out}}$ for every delay time, see Fig. 5(c). We note that $V_{\text{leak}}$ is stable in delay time with a standard deviation of only ~2.5%, indicating that a time constant of 700 ms is sufficient for the ΔF lock-in amplifier. We calculated $\Delta V_{\text{out}}$ from $V_{\text{leak}}$ using Eq. (1) and $\Delta \alpha = 0.029$ for photocurrent of 0.4 mA derived from Fig. 3(a). We corrected $V_{\text{out}}$ using the derived $\Delta V_{\text{out}}$, see the red circles in Figs. 5(c) and 5(d). Excellent agreement is achieved between the TDTR measurements at Spot B after the pump-leak correction and the conventional TDTR measurements at Spot A with a smooth surface, see Fig. 5(d).

We also employed the two-tint approach to reject the leaked pump beam in TDTR measurements at Spot B, see the blue triangles in Fig. 5(d). Excellent agreement is achieved, suggesting that both the two-tint and the proposed pump-leak correction approaches could be applied in TDTR measurements of rough samples. We note, however, that the signal-to-noise ratio of our two-tint measurements is only ~32, a factor of three smaller than our measurements using the pump-leak correction approach. The reason for this small signal-to-noise ratio is that, in the implementation of the two-tint approach, we need to increase the separation between the wavelengths of the pump and the probe beams to enhance the suppression of the high level of leaked pump beam. As a result, fluctuations in the wavelength of the laser are more significantly translated into fluctuations in the intensity of the laser beams, leading to higher noise.

**IV. LIMITS OF THE PUMP-LEAK CORRECTION APPROACH**

Here, we establish a limit for the pump-leak correction approach and compare our new approach to the two-tint method. To achieve this purpose, we performed a series of TDTR measurements on a bulk single-crystalline Cu sample coated with a 100-nm-thick Al film, with different levels of pump beam leaked into the photodiode, to mimic TDTR measurements on samples with different roughnesses. We choose the Al/Cu sample because the ratios $-V_{\text{in}}/V_{\text{out}}$ in TDTR measurements of the sample are high, and thus, TDTR measurements are more susceptible to the artifacts due to leaked pump beam. We intentionally removed the aperture in the TDTR setup and systematically changed the distance between the pump and probe beams to achieve different levels of pump leak. We quantify the leaked pump beam using the ratio $V_{\text{leak}}/V_{\text{out}}$, where $V_{\text{leak}}$ and $V_{\text{out}}$ are defined as before. For the pump power of 120 mW, the probe power of 5 mW, a spotsize of 5 μm, and a modulation frequency of 10 MHz, we achieved $V_{\text{leak}}$ ranging from 40 μV to 18 mV, $|V_{\text{out}}|$ was fixed at 150 μV, and thus $V_{\text{leak}}/V_{\text{out}}$ ranging from 0.26 to 120.

We then applied the pump-leak correction approach to the TDTR measurements acquired on the Al/Cu sample, see Fig. 6, using $\alpha = 0.033$ derived from Fig. 3(a). We find that for the case of $V_{\text{leak}} = 18$ mV, the measurements are still slightly affected by the leaked pump beam, even after the pump-leak correction. Nevertheless, we derived the thermal conductivities of Cu and found that the values are still within the range of 400 W m$^{-1}$ K$^{-1}$ ± 5%. This value of the thermal conductivity of Cu is in good agreement with the literature value.

To set a limit on the applicability of the pump-leak correction approach, we consider a safe case of $V_{\text{leak}}/|V_{\text{out}}| = 15$ ($V_{\text{leak}} = 2.2$ mV in Fig. 6). Taking into consideration the solid angle of the aperture and the reduced TDTR signals by the rough samples, we estimate that $V_{\text{leak}}/|V_{\text{out}}| = 15$ is achieved when TDTR measurements are performed on rough samples with only ~1% of the light being specularly reflected. Thus, the proposed pump-leak correction approach could be used for measurements of practically all rough samples, as long as the roughness is homogeneous. For inhomogeneously rough samples, however, laser beams are distorted at sample surfaces and thus the assumption of a Gaussian distribution of laser beams in the thermal model is violated. As a result, analysis of TDTR measurements does not yield correct results, even though the pump-leak correction approach successfully removes the artifacts due to leaked pump beam.

We summarize signal-to-noise ratios (SNR) of TDTR measurements ($-V_{\text{in}}/V_{\text{out}}$ at 100 ps) of the Al/Cu sample after the pump-leak correction approach in Fig. 7. We find that while SNR drops when $V_{\text{leak}}/|V_{\text{out}}|$ increases, a decent SNR of ~40 is still achieved when $V_{\text{leak}}/|V_{\text{out}}| = 30$, see...
Signal-to-noise ratios (SNR) of TDTR measurements on an Al/Cu sample using the pump-leak correction approach (black square) and the two-tint approach (red triangles and olive diamonds). In the implementation of the two-tint approach, we tilt the long-pass filter in the route of the pump beam such that the pump power after long-pass filter is reduced to 40% (red triangles) and 20% (olive diamonds) of the original power, respectively. We then eliminate the artifact signals from the leaked pump beam using a second bandpass filter before the photodiode. All other experimental parameters are as in Fig. 6.

![Signal-to-noise ratios (SNR) of TDTR measurements](image)

Fig. 7. For comparison, we also plot the SNR of TDTR measurements acquired using the two-tint approach. We find that using our setup, the SNR for the pump-leak correction approach is a factor of ~2 better, compared to that of the two-tint approach. The reason for the poor SNR for the two-tint approach (compared to, e.g., that reported in Ref. 16) is that our laser (from the Coherent Chameleon family), like many other ultrafast lasers, is fully automated to optimize stability in laser intensity but not in laser wavelength. As a result, fluctuations in laser wavelength are translated into fluctuations in the pump and probe powers and appear as noise in TDTR measurements.

V. CONCLUSION

In conclusion, we developed a pump-leak correction approach to eliminate the artifact signals due to the leaked pump beam in TDTR measurements of rough samples. We find that the artifact signals originate from a small phase rotation in the signals by the leaked pump beam induced by concurrent illumination of the photodetector by the modulated probe beam. The phase rotation generates an artifact signal in the out-of-phase of TDTR measurements, which can be readily reversed as long as the intensity of the leaked pump beam is monitored during TDTR measurements. Since the intensity of the leaked pump beam can be readily monitored by the AF lock-in amplifier in a typical TDTR setup, our approach does not require any additional optical elements or modification of existing TDTR setup. Also, high signal-to-noise ratios could be readily achieved even when the level of leaked pump beam is high. Our approach thus provides a convenient alternative for TDTR measurements of homogeneously rough samples.

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