A Complex Signal Fitting Method for Thermal Property Determination of TDTR Measurement

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Abstract

Time-domain thermoreflectance (TDTR) method is a widely used measurement technology for thermal property determination in micro and nanoscales. However, due to the relatively weak thermoreflectance coefficient, it is quite meaningful to improve the sensitivity and signal-to-noise ratio of the TDTR system. In general, the method of extracting thermal properties is to fit the signal (amplitude, phase or $-V_{in}/V_{out}$ signal) collected from the lock-in amplifier with higher sensitivity. In this paper, we propose a complex signal fitting (CSF) method, which directly fits the complex signal without considering the sensitivity level of the signal to the unknown parameters before solving them. As the entire output signal of the lock-in amplifier, the complex signal contains all the thermal information carried by the amplitude signal and the phase signal, so the CSF method always provides a higher sensitivity to any thermal property than the other fitting methods. Through sensitivity analysis and numerical simulation, we intuitively derive the advantage of the CSF method for various materials. To further demonstrate the validity of the CSF method, we also measured and extracted the thermal conductivity and the interfacial thermal conductance of a typical Al-SiO₂ sample with our TDTR system and CSF method. The CSF method not only simplifies the fitting process but also improves the signal-to-noise ratio of the TDTR system and the accuracy of the measured thermal properties for nanoscale materials and structures.

Keywords: Time-domain thermoreflectance; Fitting method; Sensitivity analysis; Complex signal; Thermal property.
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1. Introduction

As a relatively mature measurement method, Time-domain thermoreflectance (TDTR) is widely used in the thermal property determination of bulk and nanoscale materials due to its high spatial and time resolution, such as interfacial thermal conductance,¹³ thermal conductivity of Bas,⁴,⁵ nano-films and polymer fibers,⁶,⁷ phonon mean free path⁸,⁹ and elastic properties.¹⁰,¹¹ In a typical TDTR system, a femtosecond pulse laser is split into two beams, one is used to heat the sample, and the other one is used to monitor the temperature change with time. The principle of the TDTR system can be referred to Ref.¹²,¹³ and Fig. 1 shows the optical path schematic and the photo of the TDTR system in our laboratory. Due to the relatively weak relationship between the reflectivity and the temperature of the sample surface, the signal acquisition and data processing are challenging and important parts in TDTR measurements. To increase the sensitivity and signal-to-noise ratio of a TDTR system, a common solution is modulating the pump beam in a specific frequency and demodulating the collected signal with a lock-in amplifier to extract the small thermoreflectance signal from the strong noise environment.¹⁵,¹⁶ It can extract the amplitude of the input thermoreflectance signal as well as the phase shift between the input signal and the reference signal. In a TDTR measurement, both amplitude and phase signals contain the information of the thermal diffusion process, so fitting either of the amplitude signal or the phase signal can obtain unknown thermal properties theoretically. Other researchers have presented some improvements in the data processing such as data fitting methods and heat transfer models.¹²,¹⁷ For example, the amplitude signal fitting method is a commonly used fitting method.¹⁸,¹⁹ However, the amplitude signal involves the absorbed laser energy and the thermoreflectance coefficient which may be various for different samples, so normalization
is required in the fitting process. In 2002, Cahill et al.\cite{21} proposed a $V_{in}/V_{out}$ signal fitting method, where $V_{in}$ is the real part of the lock-in output signal and also called the in-phase signal, $V_{out}$ is the imaginary part of the lock-in output signal and also called the out-of-phase signal. This fitting method does not require normalization due to both $V_{in}$ and $V_{out}$ signals involve the laser energy and the thermoreflectance coefficient, so they are eliminated. Another similar method is the phase signal fitting method proposed by Schmidt et al.\cite{22} These two fitting methods were later widely used.\cite{12, 13, 19, 23-25}

The previous researches have shown that increasing sensitivity can improve measurement accuracy and, in some cases, can improve the efficiency of multi-parameter fitting.\cite{18, 26} In this paper, we propose a complex signal fitting (CSF) method that fits the whole complex signal from the lock-in amplifier including both $V_{in}$ and $V_{out}$ signals for the first time, and attempt to verify the effectiveness of this method by sensitivity analysis and TDTR experiment. The CSF method can provide substantial advantages over the previous fitting methods.

2. Complex Signal Fitting Method

Data fitting is the process of determining the unknown parameters by adjusting the value of the unknown thermal properties to minimize the deviation between the theoretical model and the experimental signal. In general, the method of extracting thermal properties is to fit the amplitude part, $R$, or the phase part, $\phi$, of the complex data with higher sensitivity. However, choosing which signal to fit is usually empirical. Both the amplitude signal $R$ and the phase signal $\phi$ contain the thermal property information of the sample. Usually, we choose to use the amplitude signal fitting method or phase signal fitting method to fit the data according to sensitivity analysis of different signals to the measured parameters. The fitting criterions of the two fitting methods are as follows

$$\min_{\alpha} |R_{model}(\alpha, \tau) - R_{data}(\tau)|^2$$

$$\min_{\alpha} |\phi_{model}(\alpha, \tau) - \phi_{data}(\tau)|^2$$

where $\alpha$ is the thermal property parameter to be sought. However, both $R$ and $\phi$ are only part of the output signal of the lock-in amplifier, and fitting $R$ or $\phi$ only in the data fitting process is not comprehensive. As the entire output signal of the lock-in amplifier, the complex signal combines the information of the amplitude signal and the phase signal and contains all thermal property information in the amplitude and phase signals. Therefore, we propose a complex signal fitting method, which directly fits the complex TDTR signal and provides higher sensitivity to unknown parameters. In the CSF method, we use the least squares method to fit by minimizing the square of the distance between the experimental signal and the theoretical value in the complex plane, which can be expressed as

$$\min_{\alpha} |Z_{model}(\alpha, \tau) - Z_{data}(\tau)|^2$$

where $Z = V_{in} + i \cdot V_{out}$ is the complex signal collected from the lock-in amplifier.

3. Sensitivity Analysis

3.1 Sensitivity Definition

Sensitivity analysis is an important part of TDTR experimental data processing. Whether using the amplitude signal fitting method or the phase signal fitting method, we need to perform sensitivity analysis. Sensitivity analysis can reflect the strength of the relationship between the measured signal and the extracted thermal properties for a fitting method. In the fitting process, the greater the sensitivity of the measured signal to the fitted parameter, indicating that a small change in the extracted parameter can cause a large change in the measured signal, and the more accurate results we can obtain. Therefore, through sensitivity analysis, we can choose to fit the experimental signal with higher sensitivity to the target thermal properties to get more accurate results.

A commonly used method for sensitivity definition is presented by Gundrum et al.\cite{27} They define the sensitivity of the fit $S_\alpha$ as the logarithmic derivative of signal $x$ with respect to the target parameters. Normalizing the signal can eliminate the effects of signal strength, such as the gain of instruments.

$$S_\alpha = \frac{1}{\ln(\alpha)} \frac{dx}{d\alpha} = \frac{dx}{x}$$

where $\alpha$ is the target parameter to be fitted. Aaron J. Schmidt

Fig. 1 The optical path schematic (a) and photo (b) of the TDTR system.
et al.\cite{21} extend this sensitivity definition to amplitude and phase signal fitting method for TDTR measurement according to Gundrum’s formula.

\[
S_{R,\alpha} = \frac{d \ln (R)}{d \ln (\alpha)} = \frac{dR/R}{d\alpha/\alpha} \tag{5}
\]

\[
S_{\phi,\alpha} = \frac{d \varphi}{d \ln (\alpha)} = \frac{d\varphi}{d\alpha/\alpha} \tag{6}
\]

The target parameter \( \alpha \) could be the unknown thermal parameters we are interested in, such as thermal conductivity or interfacial thermal conductance (ITC); it also could be the given parameters, such as the thickness of the metal transducer, the specific heat, the laser spot size.\cite{23} Since sensitivity is an intermediate quantity of error transmission, the smaller the sensitivity of the known parameter, the smaller the error will have on the result. So, we hope the sensitivity of the given parameters is small, and the sensitivity of the target parameters is large. Unlike the amplitude signal \( R \), the phase signal \( \varphi \) is not affected by the gain of instruments or the laser power, so there is no need to normalize the phase signal.

Here, we define the sensitivity of complex signal in a similar way in our definition, the fractional form is also adopted to eliminate the effect of system gain on sensitivity values.

\[
S_{Z,\alpha} = \frac{|dZ|/|Z|}{d\alpha/\alpha} = \frac{|dZ|/R}{d\alpha/\alpha} \tag{7}
\]

Compared with amplitude sensitivity, complex sensitivity simply replaces \( dR \) with \( |dZ| \) in the definition. However, in the following, we will see the difference between these fitting methods through sensitivity analysis.

### 3.2 Sensitivity Analysis

In a typical lock-in amplifier, the input signal and the reference signal pass through a phase-sensitive detector to form a channel, and this component is called the in-phase component. The input signal also passes through another phase-sensitive detector with a reference signal that is phase-shifted by 90° to form another channel, and this component is called out-of-phase or quadrature component. After a simple calculation of these two components, we can get the amplitude signal and phase signal we are concerned about. So, the signal \( Z \) collected from the lock-in amplifier is a complex signal, then the general form of \( Z \) is as Eq. (8), which involves all the sample properties and the TDTR system properties

\[
Z = R \cdot e^{i\varphi} \tag{8}
\]

Deriving Eq. (8), we can get

\[
dZ = d(R \cdot e^{i\varphi}) = dR \cdot e^{i\varphi} + Rd\varphi \cdot i \cdot e^{i\varphi} = dR \cdot e^{i\varphi} + Rd\varphi \cdot e^{i(\varphi + \frac{\pi}{2})} \tag{9}
\]

Converting the complex number in Eq. (9) into a trigonometric form

\[
dZ = dR \cos \varphi + i \cdot dR \sin \varphi + Rd\varphi \cos (\varphi + \frac{\pi}{2}) + i \cdot Rd\varphi \sin (\varphi + \frac{\pi}{2}) \tag{10}
\]

According to the characteristic of the trigonometric function, we can simplify Eq. (10) into the following form

\[
dZ = \sqrt{(Rd\varphi)^2 + (dR)^2} \cos (\varphi + \alpha) + i \left[ \sqrt{(Rd\varphi)^2 + (dR)^2} \sin (\varphi + \alpha) \right] \tag{11}
\]

where \( \alpha = \arctan(Rd\varphi/dR) \), then

\[
|dZ| = \sqrt{(dR)^2 + (Rd\varphi)^2} \tag{12}
\]

\[
|dZ|^2 = (dR)^2 + (Rd\varphi)^2 \tag{13}
\]

By dividing \( R^2 \) into both sides of the equation, we can get

\[
(S_{Z,\alpha} \cdot d\ln(\alpha))^2 = (S_{R,\alpha} \cdot d\ln(\alpha))^2 + (S_{\varphi,\alpha} \cdot d\ln(\alpha))^2 \tag{14}
\]

Which is

\[
S_{Z,\alpha}^2 = S_{R,\alpha}^2 + S_{\varphi,\alpha}^2 \tag{15}
\]

In the sensitivity analysis, we only care about the absolute value of the sensitivity, not concern whether it is positive or negative, so the sensitivity of the \( Z \) signal is always larger than those of \( R \) and \( \varphi \) signal.

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Fig. 2 Geometric model diagram of the TDTR complex signal, figure (b) is the partially enlarged view of figure (a).
Next, we will prove the superiority of complex sensitivity in a more intuitive way. The complex signal $Z$ collected by the lock-in amplifier is composed of the in-phase component, $V_{in}$, and the out-of-phase component, $V_{out}$, as shown in Eq. (17)

$$Z = V_{in} + i \cdot V_{out}$$

(17)

where $V_{in}, V_{out}$ is the real and imaginary part of $Z$, respectively.

For the sake of analysis, we have established the coordinate system shown in Fig. 2. According to the definition of sensitivity, the absolute value of the signal strength does not affect the sensitivity, so it can be assumed that the circle in Fig. 2 is a unit circle. $Z_1$ and $Z_2$ are the complex signals of the lock-in amplifier with two slightly different thermal properties. $Z_1$ to $Z_2$ has changed a small amount. $\phi_1, \phi_2$ are the phase angles of signals $Z_1$ and $Z_2$, respectively.

From the previous analysis, we can get

$$l_2 = dR = S_{R,\alpha} \cdot d \ln(\alpha)$$

(18)

Because $d\phi$ is a small amount, so $d\phi = l_3$, then

$$l_3 = d\phi = S_{\phi,\alpha} \cdot d \ln(\alpha)$$

(19)

According to the definition formula of complex sensitivity, we can conclude that

$$l_1 = |dZ| = S_{Z,\alpha} \cdot d \ln(\alpha)$$

(20)

Because $d\phi$ is a small amount, it can be approximated that $l_2$ is perpendicular to $l_1$.

Fig. 2(b) is a partially enlarged view of Fig. 2(a), from which we can clearly see that $l_1, l_2$ and $l_3$ are the hypotenuse and the two right angle sides of the right triangle. So, they have the following relationship

$$R^2 = I_2^2 + I_3^2$$

(21)

By dividing $[d \ln(\alpha)]^2$ on both sides of the equation, we can get the same result as Eq. (16).

We have the same conclusion as in the previous analysis, the square of the complex signal sensitivity is equal to the sum of the squares of the amplitude and the phase signal sensitivities, so $S_{Z,\alpha}$ is always greater than $S_{R,\alpha}$ and $S_{\phi,\alpha}$.

4. Advantages of Complex Signal Fitting Method

To demonstrate the advantages of the CSF method, firstly, we calculated the parameter sensitivities for two typical samples, Al coated silicon dioxide (SiO$_2$) and diamond samples. The silicon dioxide represents materials with low thermal conductivities, while the diamond represents materials with high thermal conductivities. Fig. 3 shows the sensitivities of the thermal conductivity of the substrates and the ITC between Al transducer and substrates calculated with a modulation frequency of 5 MHz. The blue, red and yellow lines represent the sensitivities of amplitude signal, phase signal and complex signal, respectively. The green dotted lines represent the maximum delay time of our TDTR experimental system, 7 ns.

Fig. 3 Simulated sensitivities of the thermal conductivity and the ITC for silicon dioxide and diamond samples. (Higher is better.) Figure (a) and (b) are the sensitivities of the thermal conductivity of SiO$_2$ and ITC between Al and SiO$_2$, figure (c) and (d) are the sensitivities of the thermal conductivity of diamond and ITC between Al and diamond.
As shown in Fig. 3(a) and 3(c), as the delay time increases, the amplitude sensitivity and phase sensitivity gradually intersect, which means neither of the two signals is always more sensitive than the other one. The phase sensitivity is greater when the delay time is small, but the opposite is true when the delay time is large. If the experimental data is fitted according to the previous fitting method, the experimental signal to be fitted should be selected according to the specific situation. For example, within the maximum delay time of 7.5 ns in our experimental system, the amplitude sensitivity in Fig. 3(a) is greater than the phase sensitivity most of the time, however Fig. 3(c) is the opposite. So, for our experimental system, the amplitude signal should be selected for fitting the thermal conductivity of SiO$_2$, while the phase signal should be selected when fitting the thermal conductivity of diamond. However, the complex sensitivity is always greater than the amplitude and phase sensitivities over the full delay time. Therefore, whether it is fitting the thermal conductivity of SiO$_2$ or diamond, the CSF method should be a better choice.

The situations in Fig. 3(b) and Fig. 3(d) are different from the previous description. As shown in the figure, both the ITC of the Al-SiO$_2$ sample and the ITC of the Al-diamond sample are more sensitive to the amplitude signal than the phase signal throughout the delay time. The sensitivity of ITC to the complex signal is still the largest among the three signals, but the sensitivity of amplitude signals is close to that of the complex signal. In this case, fitting the amplitude signal or the complex signal is almost the same. Here, we choose 0.5 ns as the starting delay time of the fitting, and the data at 0.5 ns should be the baseline. However, to eliminate the influence of random system noise, eleven data around 0.5 ns are averaged as the baseline. Then we normalized the complex data by dividing them by the amplitude of the baseline, and the normalization process does not change the phase component of the complex signal. However, choosing a different delay time as the baseline will affect the fitting results. Fig. 4 shows the fitting results obtained under different normalization time. It can be seen that after 0.5 ns, the fitting result tends to be stable, therefore, we chose 0.5 ns as the baseline in this case. The figure also shows that the amplitude signal fitting, which also needs normalization, are more fluctuating than the complex signal fitting when the normalization is performed at different delay time.

![Fig. 4 Thermal conductivity fitting results under different normalization delay time. The blue line and the red line represent the complex signal fitting and amplitude signal fitting, respectively.](image)

To demonstrate the fitting process and feasibility of the CSF method, we also prepared an Al-SiO$_2$ sample and measured the thermal conductivity and the ITC using our TDTR experimental system. The $V_{\text{in}}$ and $V_{\text{out}}$ signals are first collected from the lock-in amplifier and then constructed to complex numbers. Since the strength of the complex signal will be affected by the absorbed laser energy, the thermal reflectance coefficient, and the electronic gains, etc., we choose the data at a specific delay time as a baseline to normalize the complex signal, just like the amplitude signal. The situations in Fig. 3(b) and Fig. 3(d) are different from the previous description. As shown in the figure, both the ITC of the Al-SiO$_2$ sample and the ITC of the Al-diamond sample are more sensitive to the amplitude signal than the phase signal throughout the delay time. The sensitivity of ITC to the complex signal is still the largest among the three signals, but the sensitivity of amplitude signals is close to that of the complex signal. In this case, fitting the amplitude signal or the complex signal is almost the same.

![Fig. 5 TDTR experimental data and the best fitting curves by the CSF method. The blue and red lines are in-phase signal and out-of-phase signal collected from the lock-in amplifier, the yellow and purple lines are the best fitting curves.](image)

The complex signal of the experimental data and the best fitting curves are shown in Fig. 5. It can be seen from the figure that $V_{\text{in}}$ and $V_{\text{out}}$ components given by the CSF method fit well with the experimental data, the extracted ITC between Al and SiO$_2$ is ~90 MW·m$^{-2}$·K$^{-1}$, and the thermal conductivity of SiO$_2$ is 1.35 W·m$^{-1}$·K$^{-1}$, which is consistent with the literature values. In this case, the sensitivity of the complex signal is close to the amplitude signal, and the CSF results are also found to be close to the amplitude fitting results as expected.

5. Conclusion
We proposed a new complex signal fitting (CSF) method for TDTR experimental data processing, which combines the amplitude signal fitting method and the phase signal fitting method. We proved that the CSF method always provides a higher sensitivity to any parameter in theoretical analysis and numerical simulation, and illustrated the feasibility of the CSF method by experiments. However, the increased sensitivity of given parameters might make the CSF method more uncertain than the other fitting methods in some cases, which needs to...
be determined according to the specific experimental conditions and sample structures. Still, we think it’s a better choice to utilize a fitting method with higher sensitivity. This method will improve the signal-to-noise ratio of the TDTR system and the accuracy of thermal transport measurement for nanoscale materials and applications.

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Supporting information
applicable

Conflict of interest
There are no conflicts to declare.

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