Thermal characterization of Au-Si multilayer using 3-omega method

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Abstract

As thermal management becomes a serious issue in applications of thermoelectrics, thermal insulation and etc., understanding of heat transport in highly low thermal conductive system has been required. Here, 3-omega technique to measure thermal conductivity is demonstrated, where black-body radiation losses are significantly reduced by moving the measurement into frequency domain so that it expands the experimental reliability to measure ultralow thermal conductivity. Also, there is an experimental example of thermal characterization of Au-Si multilayers using this measurement.

1. Background of 3-omega method

A conventional steady-state system to measure the thermal conductivity is to apply power source to one side of a sample. The temperature profile within the sample can be considered as quasi-one-dimensional near the center. To extrapolate the thermal conductivity, Fourier's Law of heat conduction is used as:

$$k = -\frac{Q \cdot t}{A \cdot \Delta T} \qquad (1)$$

where Q is the input power, ΔT is the normal temperature difference, and t and A are the thickness and cross-sectional area of the specimen, respectively. However, the experimental difficulties begin with the limited capabilities for measuring thermal transport in thermally low conductive materials (i.e. glass) and/or thermal conductivity measurement at high temperatures. This is mainly due to long thermal equilibrium times and to errors caused by blackbody infrared radiation [1].

The 3-omega (3ω) technique increases the accuracy in measuring thermal conductivity by moving the measurement into the frequency domain. To be specific, by applying an AC current with a frequency of ω, one can measure the voltage with a frequency of 3ω, which measured information enables to extract the value of thermal conductivity. As shown in Fig. 1, this method utilizes a patterned metal line deposited on the samples both as a heater for applying a periodic heat flux and a sensitive thermometer for measuring the surface temperature [2]. By applying an AC electrical current (I(ω)) modulated at an angular frequency ω to the metal line, the periodic heating results in a heat flux (P) oscillating at 2ω as:

$$P = \left[I_0 \sin(\omega t) \right]^2 R = \frac{I_0^2 R}{2} + \frac{I_0^2 R}{2} \cos(2\omega t) \qquad (2)$$

where I_0 is the current amplitude, ω is the applied frequency, R is the resistance of the metal strip and P is the heating power. The power consists of two components: a constant component independent of time (first term in Eq. 2) and an oscillating component (second term in Eq. 2). Because the temperature rise in the metal strip is directly proportional to the heat flux, it also has a component modulated at 2ω with a phase shift ϕ.

$$T_{rise} = T_{DC} + T_{2\omega} \cos(2\omega t + \phi) \qquad (3)$$

which leads to variations in the resistance given by Eq. 4 since the resistance of the metal strip is dependent on the temperature, which property depends on the materials and it is called temperature coefficient of resistance (TCR).

$$R_{total} = T + \frac{dR}{dT} T_{DC} + \frac{dR}{dT} T_{2\omega} \cos(2\omega t + \phi) \qquad (4)$$

Finally, the resulting voltage across the metal heater line is obtained by:

$$V_{3\omega} = \frac{I_0}{2} \frac{dR}{dT} T_{2\omega} \sin(3\omega t + \phi) \qquad (5)$$

where $T_{2\omega}$ is the second harmonic of the temperature rise, which depends on the thermal conductivity of the materials. Therefore, by extracting $T_{2\omega}$ from the measured $V_{3\omega}$ signal according, the thermal conductivity of the sample can be obtained.

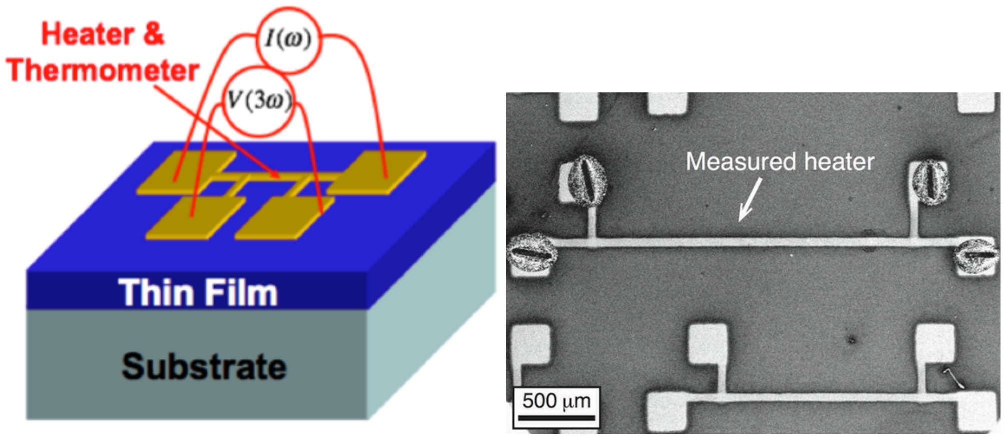

Fig. 1 Schematic diagram (left) and (a scanning electron microscopic (SEM) image (right) of a device for 3ω measurement for thin film sample [refers 3 and 4].

2. Ultralow thermal conductivity of multilayers

The interfacial thermal resistance plays a significant role in transporting of thermal energy in nano-scale structures, where their scale is similar to or less than the mean free path of phonon (about 10-100 nm) because the interface limits the transport of phonon [5]. For an interface between dissimilar materials, the different densities and sound speeds result in a mismatch in the acoustic impedances [6]. Based on the Eq. (6), it implies that the high interfacial thermal resistances are composed of high (hard) and low (soft) Debye temperature materials.

$$\Theta_D = \frac{\hbar\omega_D}{k_B}\frac{V}{6\pi^2}\left(\frac{1}{\upsilon_L^3}+\frac{2}{\upsilon_T^3}\right)\omega_D^3 \qquad (6)$$

where Θ_D is Debye temperature, ω_D is Debye cut-off frequency, k_B is Boltzmann constant, υ_L is longitudinal sound velocity, υ_T is transverse sound velocity and V is volume [5]. The sound velocity is generally determined by Newton-Laplace equation ($\upsilon = \sqrt{K/\rho}$, where K and ρ are a coefficient of stiffness and the density, respectively).

Therefore, the relation between the interface thermal resistance can be characterized by the ratio of Debye temperature.

Dechaumphai and co-workers studied on ultralow thermal conductivity of multilayers, consisting of Si and Au that have highly dissimilar Debye temperatures (i.e. Si and Au have Debye temperature of 645 and 162, respectively [7]). They experimentally showed a thermal conductivity of 0.33 ± 0.04 W m^{-1} K^{-1} at room temperature with the interfacial density of ~0.2 interface nm^{-1}[4]. As described in Fig. 2, the entire thermal resistance can be represented by a series of each element, including resistance of Au, Si and their interfaces. Therefore, the thermal resistance of one element (R_{unit}), consisting of a layer of Au (R_{Au}), Si (R_{Si}) and interfaces ($2R_{pp}$) can be given by:

$$\mathrm{R}_{unit} = R_{Si} + R_{Au} + 2R_{pp} \qquad (7)$$

where the R_{unit} can be extracted from the experimentally measured thermal conductivity (=d/R_{unit}, where d is thickness of one element), and the experimental results of thermal conductivity is discussed in following section 3. Subtracting the values of R_{Si} and R_{Au} from R_{unit}, R_{unit} was obtained, and it was found that a contribution of interface on the entire thermal resistance exceeds ~60% in multilayers of Au and Si [4].

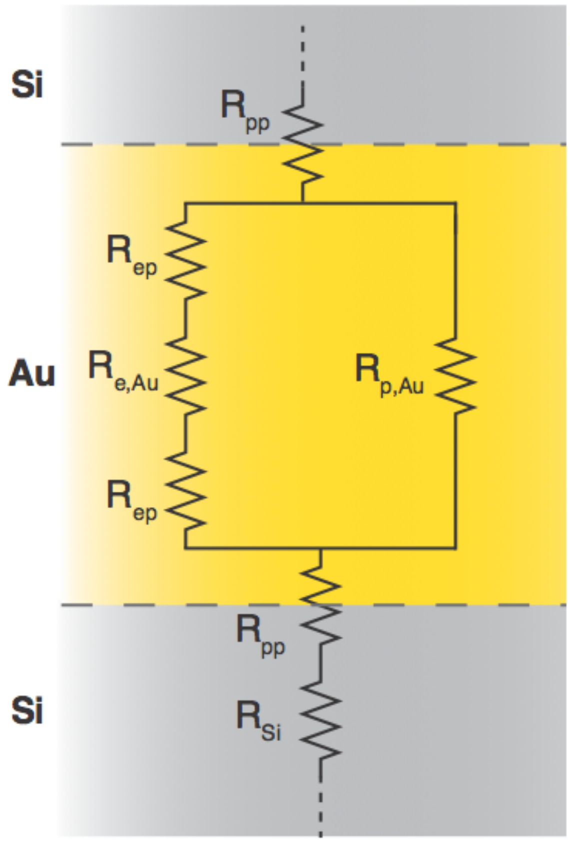

Fig. 2 Schematic of equivalent circuit of thermal resistance in multilayers of Au and Si, where the each element (i.e., R_{Si}, R_{Au} and R_{pp}) is serially connected [refer 4].

3. Measurement of thermal conductivity using 3-omega method

To elucidate thermal conductivity, firstly temperature rise should be measured as a function of frequency (see Fig. 3). Since the 3ω method gives the information on the entire layer, including substrate and multilayers, the reference sample without multilayers was measured to extract the only information on multilayers. Based on the geometry and thermal properties of targeting samples, the relationship between thermal conductivity and measured temperature can be established. In this film structure where deposited heater width is much larger than the thickness of sample, two dimensional heat conduction model can be employed.

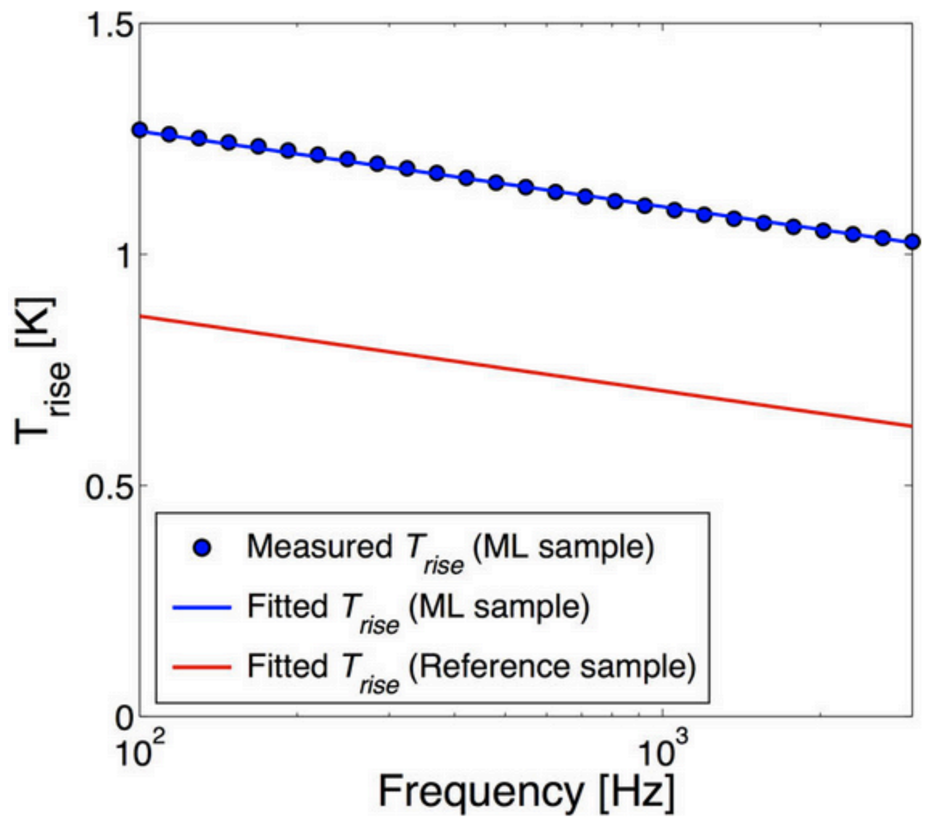

Fig. 3 Experimental temperature rise with fitting results as a function of frequency measured by 3ω method [refer 4].

As shown in Fig. 4, the obtained thermal conductivity of 0.33 ± 0.04 W m^{-1} K^{-2} at room temperature is even lower than amorphous limit (i.e. 1.05-1.6 W m^{-1} K^{-2} for Si and 0.49 W m^{-1} K^{-2} for Au) due to the highly dissimilar interfaces [4]. Also, to confirm the influence of interfaces on the entire thermal conductivity, Dechaumphai et al. compared the data with the data of samples after annealing process to make the interface ambiguous. As expected, annealed samples showed increased thermal conductivity (1.06 ± 0.20 W m^{-}

[1] K^{-2} at room temperature) due to the lack of distinct interfaces [4]. It confirms that the interfaces between Au and Si block the transport of phonon.

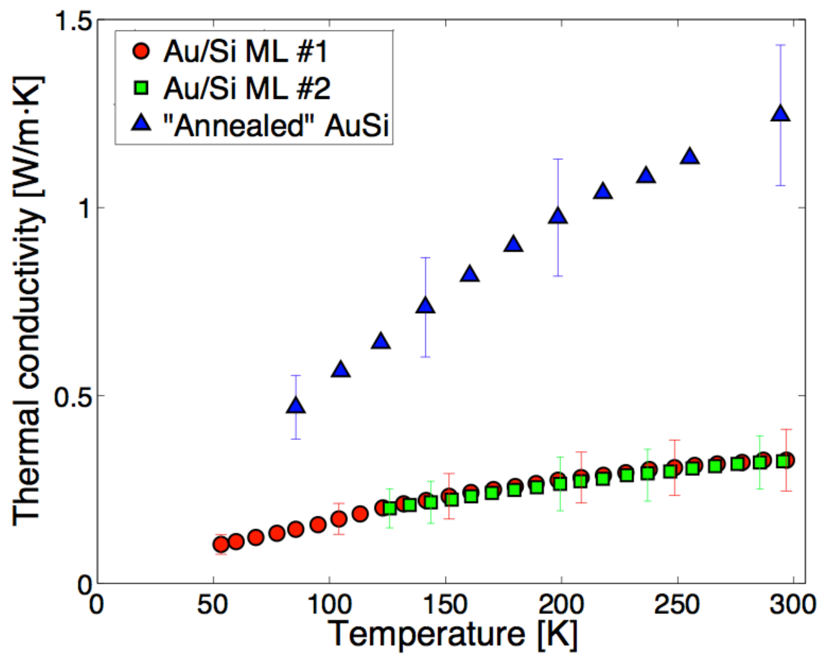

Fig. 4 Measured thermal conductivity as a function of temperature, where the measurement was conducted in vacuum chamber. Samples before (circle) and after (triangle) annealing process was compared [refer 4].

4. Conclusion

In addition to the advantages of 3ω method, such as minimizing radiation loss and reducing thermal equilibration times, the frequency dependent property makes the measurement system more sensitive with the help of lock-in amplifier. Therefore, the 3ω technique is an excellent candidate in that an ultralow (even lower than amorphous limit) thermal conductivity can be measured. This technique enables to study on understanding thermal transport in nanostructured and/or amorphous materials.

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