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Influence of upper layer on measuring thermal conductivity of multilayer thin films using differential 3-ω method

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ABSTRACT

We present an intrinsic error in differential 3-ω measurement method due to two-dimensional heat spreading effect in the upper layer of the target film. By measuring thermal conductivities of 300 nm PECVD-grown silicon dioxide thin films with various thicknesses of upper layers, significant heat spreading effect is observed. Also, analytical modeling regarding apparent thin film thermal conductivity is conducted for verification of experimental results. Experimental results as well as analytical results show that the measurement error tends to increase with thickness of upper layer due to two-dimensional heat spreading effect.

1. Introduction

Since D. G. Cahill developed the well-known 3-ω method for thermal conductivity measurement [1], numerous researches not only about thin film thermal transport [2–6] but also methodology itself [7–10] have exploited briskly. The 3-ω method, which is based on two-dimensional heat diffusion for bulk materials (slope method) and one-dimensional heat conduction for thin films (differential method), is an AC modulating technique for measuring thermal properties of planar materials [1,2]. Due to its wide adaptability and reliability, thermal transport in various materials such as superlattices [3–5], nanowire arrays [11], micro/mesoporous thin films [12] are of great interest in recent. However, since 3-ω method is based on several assumptions, there exist few preconditions that must be satisfied in order to extract precise results such as line heat source approximation, semi-infinite medium and one-dimensional heat conduction in the target film [7].

For some cases, additional layers must be included at the top and/or bottom of the target film as an insulator, stabilizer, passivation layer, or buffer layer. When measuring thermal conductivity of a multilayer sample, heat spreading in not only target film but also additional layers must be removed for satisfying one-dimensional heat conduction approximation. However, these additional layers can be inevitably thick for some cases such as polymer coatings for nanowire arrays [11], buffer layers for mesoporous thin films [12], which results in an undesirable two-dimensional heat spreading. Fig. 1 shows a concept of two-dimensional heat spreading in multilayer thin films. Heat spreading in additional layers can induce crucial error although the target film has satisfied one-dimensional heat conduction condition.

In this paper, we present effect of additional upper layer on differential 3-ω thermal conductivity measurement. Also, theoretical modeling regarding apparent thermal conductivities of consisting layers is conducted for extensive analysis.

2. Experimental details

The 3-ω thermal conductivity measurement method originates from conventional hot-wire technique. A thin metal wire is employed at the top of the sample for simultaneous operation as a heater and a thermometer. An alternating input current with frequency ω heats the sample with 2ω frequency. Since electrical conductivity of metal varies linearly with temperature, output voltage includes 3ω component which depends on thermal conductivity of the sample. The 3ω voltage signal can be extracted using lock-in amplifier.

Fig. 2 shows schematic of experimental sample sets. Results were given by comparing thermal conductivity of single 300 nm SiO_2 thin film with same film including SiN_x upper layer with thicknesses of 60, 300, and 2000 nm. SiO_2 and SiN_x thin films were grown by PECVD (Plasma enhanced chemical vapor deposition) process. 5%SiH_4/N_2, N_2O, and N_2 gases with flow rate of 160 sccm, 1500 sccm, and 240 sccm, respectively, were used to deposit SiO_2 thin film under ambient pressure of 73.3 Pa. For SiN_x thin film deposition, 5%SiH_4/N_2, NH_3, and N_2 gases with flow rate of 800 sccm, 10 sccm, and 1200 sccm were used under ambient pressure of 77.3 Pa. Deposition temperatures were 573 K for both films. After PECVD deposition, 10 nm Cr adhesion layer followed by 300 nm Au thin film were deposited using

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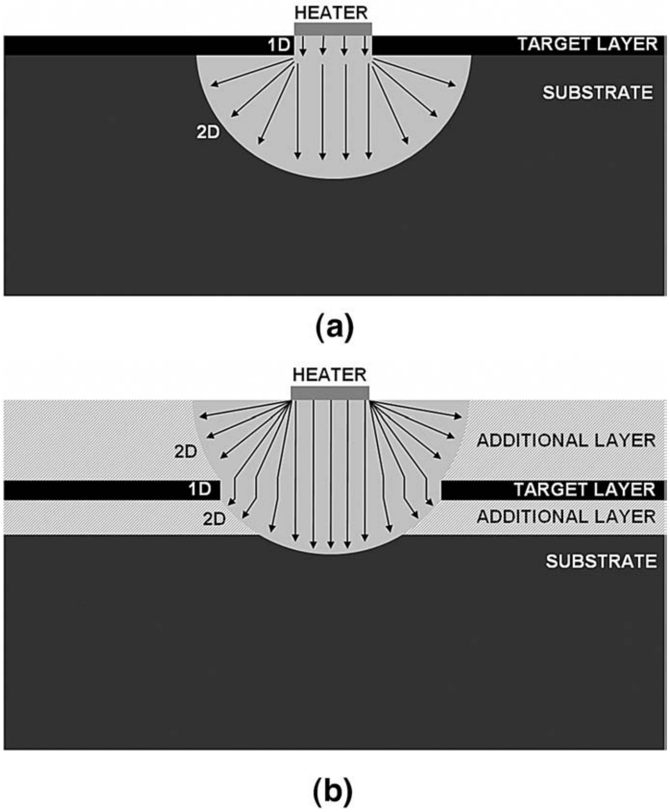

Fig. 1. Schematics of heat flow in thin film samples. (a) General heat flow in 3-ω method sample. (b) Undesirable two-dimensional heat spreading in additional layers.

e-beam evaporation. The Au heater, which has geometries of 12 μm in width and 1.5 mm in length, was patterned by lift-off technique. Thermal conductivity measurements were carried out at room temperature (300 K) under vacuum condition (4×10^{-4} Pa) for minimizing convective heat loss to ambient.

3. Analytical modeling

For analytical modeling, two-dimensional heat diffusion equation was employed for anisotropic multilayer films which were derived using integral Fourier transform technique [7,8]. The complex temperature rise of heater with power dissipating per unit length is as follows.

$$\Delta T = \frac{-p}{\pi l k_{y_1}}\int_0^\infty \frac{1}{A_1B_1}\frac{\sin^2(bq)}{b^2q^2}dq \tag{1}$$

where A_1 can be determined by

$$A_j = \frac{A_{j+1}\frac{k_{y_{j+1}}B_{j+1}}{k_{y_j}B_j} - \tanh(\varphi_j)}{1 - A_{j+1}\frac{k_{y_{j+1}}B_{j+1}}{k_{y_j}B_j}\tanh(\varphi_j)} \quad j = 1, 2, \ldots, n-1 \tag{2}$$

and B_1 can be determined by

$$B_j = \left(k_{xy}q^2 + \frac{i2\omega}{\alpha_{y_j}}\right)^{1/2} \quad j = 1, 2, \ldots, n-1 \tag{3}$$

and

$$\varphi_j = B_j d_j, \quad k_{xy} = k_x/k_y \tag{4}$$

Here, p is the input power and ω is the input frequency. l and b are length and width of heater line respectively. n is the total number of

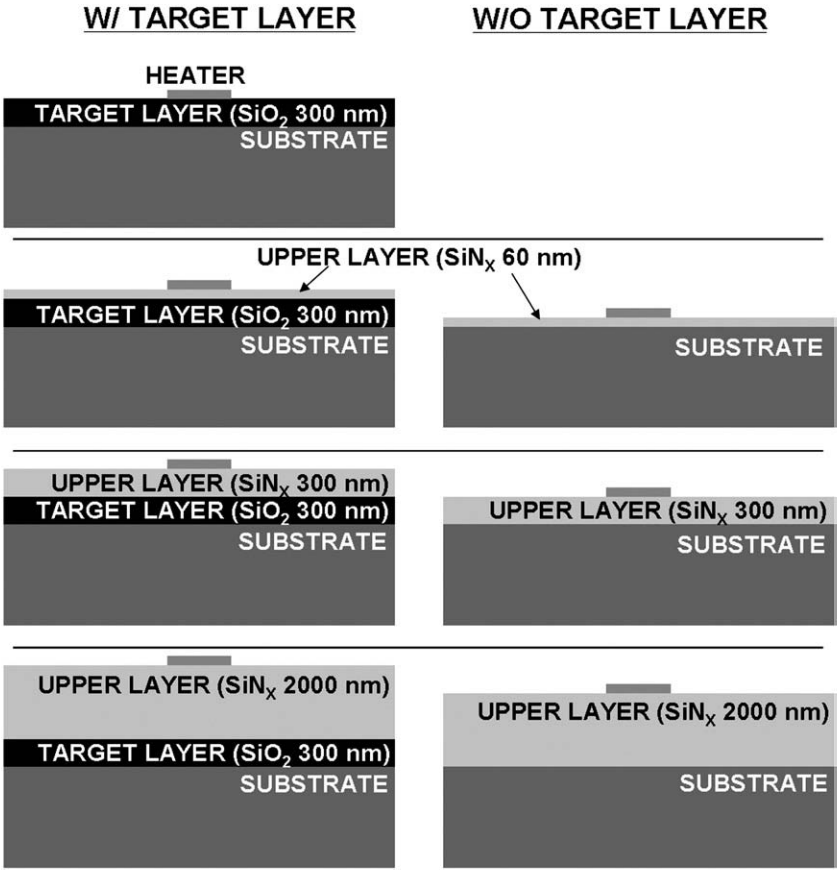

Fig. 2. Schematics of experimental samples.

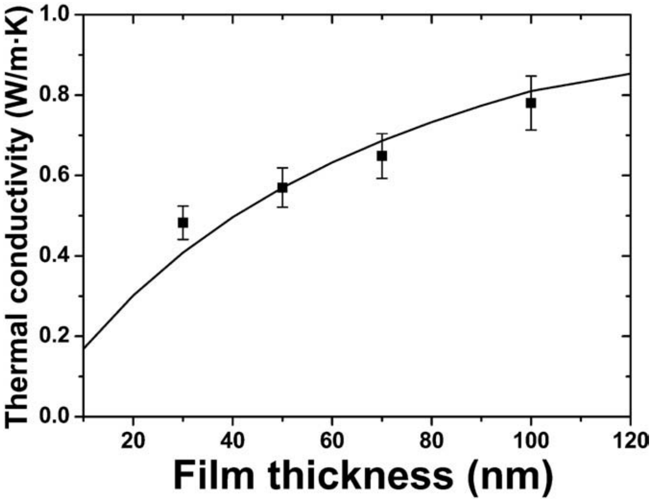

Fig. 3. Thermal conductivity of amorphous SiN_x thin films with film thickness. Uncertainty of the experiment was determined to be 8.6% at the confidence level of 95% [13]. The solid line represents fit to Eq. (5).

film layers beginning with the first film under the metal heater line ($j=1$) through the semi-infinite substrate ($j=n$) of thickness d_n. Also, for semi-infinite condition, $A_n=-1$. α_y refers to the thermal diffusivity in the cross-plane direction. The Fourier space variable, q, is used for the integration. Since SiO_2 and SiN_x thin films have isotropic transport behavior, k_{xy} is set to unity.

For amorphous materials, intrinsic thermal conductivity is independent of film thickness because of short mean free path of dominant heat carriers which is in order of several Angstroms to few nanometers at room temperature. However, for thin films with thickness of submicron, finite thermal boundary resistance becomes increasingly important as film thickness decreases. Therefore, thermal conductivities in Eq. (1) must be treated as apparent thermal conductivities. Apparent thermal conductivity can be derived using Matthiessen's rule about mean free path of heat carriers [2].

$$k_a = \frac{k_{\mathrm{i}}}{1 + R_{\mathrm{I}} k_{\mathrm{i}}/t} \tag{5}$$

where k_{i} is the intrinsic thermal conductivity which is independent of its size, R_{I} is thermal boundary resistance, and t is film thickness. Fig. 3 shows measured apparent thermal conductivity of PECVD SiN_x films with various film thicknesses at 300 K. Experimental uncertainty, which was suggested by Kline and McClintock [13], was calculated as 8.6% at confidence level of 95%. By fitting Eq. (5) to the experimental results, thermal boundary resistance was estimated to be $R_{\mathrm{I}} \sim 5.2 \times 10^{-8}$ m^2 K/W which is slightly larger but comparable to previous studies [2,8]. The discrepancy may have caused by existing impurities and/or contaminations located at the boundaries.

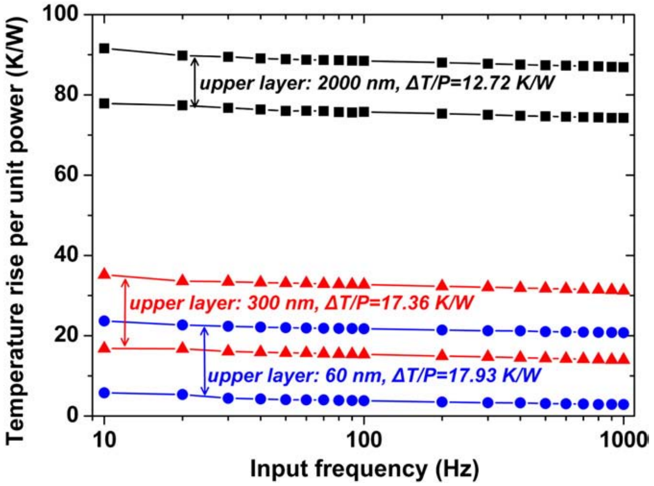

Fig. 4. Temperature rise of experimental samples at heater line with respect to input current frequency.

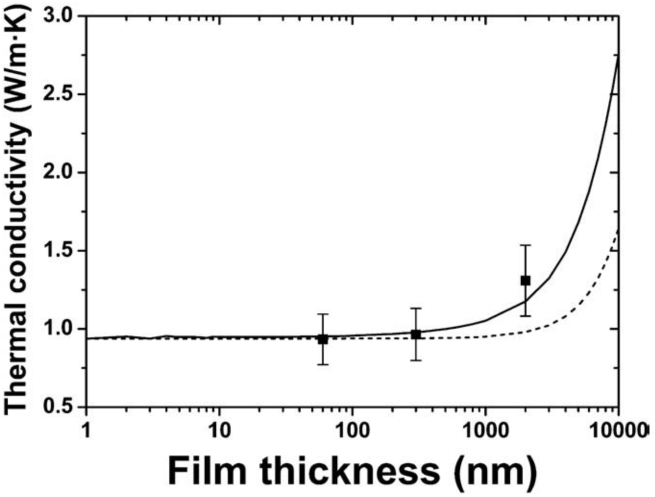

Fig. 5. Thermal conductivity with varying thicknesses of upper layers. Uncertainty of the experiment was determined to be 17.3% at the confidence level of 95% [13]. The solid line represents predicted values regarding apparent thermal conductivity of consisting layers. The dashed line represents the same values regarding intrinsic thermal conductivities of each layers.

4. Results and discussions

Fig. 4 shows typical temperature rise per unit power at the heater lines of the experimental samples with respect to input current frequency. In Fig. 5, the measured thermal conductivities are compared to analytically calculated thermal conductivity using Eq. (1). In detail, this figure represents thermal conductivity of 300 nm SiO_2 thin film with various thicknesses of SiN_x upper layers. The solid line indicates calculated thermal conductivity using Eq. (1) regarding apparent thermal conductivities while the dashed line indicates the

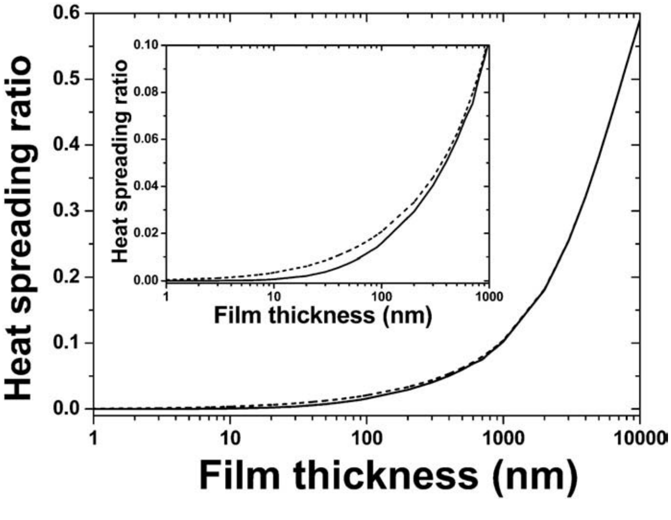

Fig. 6. Heat spreading ratio in upper layer. The solid line represents calculation regarding apparent thermal conductivity while the dashed line represents same values regarding intrinsic thermal conductivities. The inset shows magnified results for clear observation of disparity between two different calculations.

same values regarding intrinsic thermal conductivities of each layers. In other words, solid line deals thermal conductivity and thickness of each layer linked together while dashed line deals such parameters independently. Analytical result considering apparent thermal conductivity is consistent with experimental values because it reflects actual conditions.

From Fig. 5, the measured thermal conductivity tends to increase with thickness of upper layer, which is purely an experimental error. The measured thermal conductivity of single layer of 300 nm SiO_2 thin film was about 0.94 W/m K. Also, the same layers with upper additional layer of 60 nm or 300 nm SiN_x were similar to such value, which were about 0.93 W/m K and 0.96 W/m K, respectively. However, as the thickness of upper layer exceeds order of few microns, disparity between measured value (w/ upper layer) and true value (w/o upper layer) tends to increase dramatically. With SiN_x upper additional layer of 2 μm thickness, about 40% higher value (1.31 W/m K) was obtained compared to the single layer value. Experimental uncertainties were about 17.3% at confidence level of 95% [13]. The reason being is that the two-dimensional heat spreading effect has occurred in the upper layer. As thickness of the upper layer increases, thermal wave loses most of its energy in the upper layer and the target film may not be recognized by incoming thermal wave. In other words, although target film may have satisfied several preconditions which were mentioned above [7], thermal resistance of the whole sample is dominated by the upper layer which results in dramatic decrease of experimental accuracy. Therefore difference of temperature drop between the target sample, which consists of target layer (SiO_2) and upper layer (SiN_x), and the reference sample, which consists of upper layer only, becomes smaller. Owing to one-dimensional heat conduction equation, such result brings larger thermal conductivity in consequence.

Additionally, heat spreading ratio in the upper layer is presented in Fig. 6. This value represents the ratio of heat generated from the heater to heat traveled sideways (in-plane direction) inside the upper layer, where zero being no heat spreading. Obviously heat spreading ratio increases with thickness of upper layer, which is consistent with our analysis. Also, it can be clearly seen that the amount of heat spreading occurred in the upper layer for calculation with apparent thermal conductivity (solid line) and that with intrinsic thermal conductivity (dashed line) are different. This difference will be increased when the thermal conductivity contrast between the substrate and consisting layers decreases, which once again emphasizes the importance of actual thermal properties in thermal analysis.

It should be pointed out that the effects of thermal mass of heater and heater width on two-dimensional heat spreading in thin films have not been considered in our analysis. However, these factors are negligible for low input frequencies. Also, it is obvious that the thickness is the most dominant factor that influences heat spreading effect, and thus should be considered in the first place. Moreover, thermal conductivity must be modified for submicron films since it is dependent on the film thickness.

5. Conclusion

In summary, experimental as well as analytical results presented in this work show that the measurement error in differential 3-ω measurement is induced by increasing thickness of upper additional layer due to the basis of its technique. One-dimensional heat conduction cannot be applied when the thickness of upper layer exceeds certain amount, which in this work, 360 nm for experimental results satisfying less than 5% of error. Furthermore, actual properties and conditions must be taken into account for accurate thermal modeling and analysis when dealing with materials those intrinsic properties cannot hold anymore.

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