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A Simple Efficient Method of Nanofilm-on-Bulk-Substrate Thermal Conductivity Measurement Using Raman Thermometry

Vladimir Poborchii[1], Noriyuki Uchida[1], Yoshinobu Miyazaki[1], Tetsuya Tada [1], and Pavel Geshev [2]

[1] Nanoelectronics Research Institute, National Institute of Advanced Industrial Science and Technology, 1-1-1 Higashi, AIST Central-5, Tsukuba 305-8565, Japan
Phone: +81-298-491633, e-mail: vladimir.p@aist.go.jp
[2] Institute of Thermophysics of the Russian Academy of Sciences, Lavrentyev Ave. 1, and Novosibirsk State University, Pirogova Str. 2, Novosibirsk 630090, Russia

Abstract

Using Raman thermometry, we established a simple efficient method of thermal conductivity (*k*) measurement for light-absorbing nanofilms on bulk non-absorbing substrates with low *k*. We determine film *k* comparing experimental and theoretical laser-induced film heating calculated assuming diffusive phonon transport (DPT). We show that the method works for films with thickness *h* larger than the phonon mean free path *Λ*, even for low *k* materials like nanocrystalline Si. For *h* < *Λ*, despite ballistic phonon transport contribution, this approach works when the in-plane DPT dominates, e.g. in Si films on quartz with *h* ≥ 60 nm. Our method is simpler and more efficient than any other existing NF *k* measurement method, a dozen of NFs being examined in one hour.

1. Introduction

Measurement of the thermal conductivity (*k*) of nanofilms (NFs) and, especially, Si-based NFs is an important task for science and industry. Bulk Si was never used in thermoelectric applications due to its high *k* ~ 150 W/m/K and, therefore, low thermoelectric figure of merit *ZT* < 0.01 at room temperature. A reduction of *k* in Si NFs and nanowires (NWs) due to phonon boundary scattering and other effects can improve their performance. Further *k* reduction can be achieved in nanocrystalline Si-based NFs, which are composites of Si and Ni silicide nanocrystals (Ni-Si NC). Due to both phonon scattering at the grain boundaries and planar defects, the films have much lower *k* (3 – 7.7 W/m/K) and higher *ZT* (> 0.1) than bulk Si at room temperature [1]. In this work, we examine *k* in Si NFs and composite NC NFs of Si and Fe silicide (Fe-Si NC) using Raman thermometry. Recently, Raman scattering was utilized to measure *k* in nano-membranes (NMs) [2] and suspended NWs [3]. Situation is more complex for NFs on bulk substrates playing a role of the heat drain. Raman spectroscopic measurement of the *k* of such NFs is still a challenge. In this work, we solve this problem for the case of absorbing NF on bulk non-absorbing low-*k* substrate and measure *k* of Si NFs and composite Fe-Si NC NFs on quartz.

2. Theoretical and Experimental Methods

Let us consider an absorbing layer with complex refractive index $N_1 = \sqrt{\varepsilon_1} = n_1 + i\kappa_1$ on an infinite substrate with the refractive index $N_2 = \sqrt{\varepsilon_2} = n_2$. Laser light hits the absorbing layer from media with the refractive index $N_0 = \sqrt{\varepsilon_0} = n_0$. Density of the light power flux is

$$S = (P/\pi r_0^2)\exp(-r^2/r_0^2) \qquad (1)$$

where *P* is the laser power while r_0 is the radius of the focused laser light beam (Fig. 1).

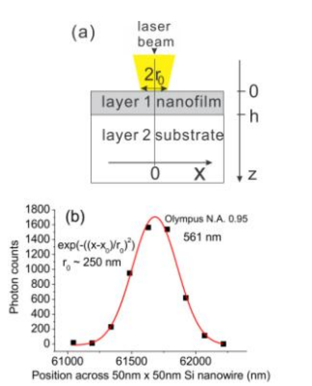

Fig. 1. a) Laser illumination of absorbing NF on transparent substrate; b) Measured light intensity distribution in the focused 561 nm laser beam (black squares) and its Gaussian fitting (red curve)

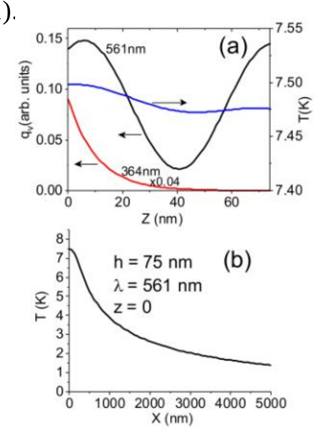

Fig. 2. a) Volumetric heat density $q_V(z)$ at *x*=0 in 75 nm thick SOQ by the 561 nm (black curve) and 364 nm (red curve) light absorption; the temperature field *T(z)* (blue curve) for the 561 nm light with *P* = 1 mW; b) Temperature field *T(x)* at z=0 for 75 nm thick SOQ.

The light is propagating from the semi-space 0 (z < 0), hitting the absorbing layer 1 (0 ≤ *z* ≤ *h*) and propagating further into semi-space 2 (*z* > *h*). Volume density of heat generated by the light absorption can be expressed as

$$q_V = \varepsilon_1'' |E_1|^2 \omega/(8\pi) \qquad (2)$$

where ω is the light frequency, ε_1'' is the imaginary part of dielectric constant while E_1 is the electric field of light in the layer 1. We calculate E_1 first.Then we solve stationary heat transfer problem for layered medium with temperatures T_1 and T_2 thermal conductivities k_1 and k_2 in layers 1 and 2, respectively. The governing equations are

$$\left(\frac{\partial^2}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r} r \frac{\partial}{\partial r}\right) T_1 = -\frac{q_V}{k_1} \qquad 0 \le z \le h \qquad (3)$$

$$\left(\frac{\partial^2}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r} r \frac{\partial}{\partial r}\right) T_2 = 0 \qquad (z \ge h) \qquad (4)$$

For $z=0$ $\frac{\partial T_1}{\partial z'}=0$;for $z=h$ $T_1=T_2$ $k_1\frac{\partial T_1}{\partial z'}=k_2\frac{\partial T_2}{\partial z'}$

At $r\rightarrow\infty$ $T_1=T_2=0$; at $z\rightarrow\infty$ $T_2=0$

After Hankel transform, we obtain a system of equations that we solve using Green's function method. Finally, we calculate an averaged laser-induced heating ΔT_{calc} of the illuminated area of layer1.

$$\Delta T_{calc}=\int_{V_1}T_1(\vec{x})q_V(\vec{x})d^3\vec{x}\,/\int_{V_1}q_V(\vec{x})d^3\vec{x} \qquad (5)$$

Raman measurement was done using a Nanofinder 30 confocal Raman system (Tokyo Instruments Inc.) equipped with a 561 nm wavelength laser. Lens with 100× magnification and 0.95 numerical aperture was used to focus laser light onto spot with r_0, ~ 250 nm. ΔT was determined from the laser-induced Raman downshift $\Delta\omega$.

For testing our method, we measured k of Si on quartz (SOQ) and Si on sapphire (SOS) NFs. Their thinning was done by thermal oxidation with subsequent etching in HF. Composite $FeSi_{20}$-2P NFs were fabricated using phase separation from amorphous Fe–Si alloy films that had a composition ratio of Si/Fe ~ 20 similar to Ni-Si [1]. The amorphous films were deposited on fused silica substrates by DC magnetron sputtering of a target. The sputter deposition was carried out in pure Ar gas at a sputtering pressure of 3.0×10^{-3} Torr. The thickness of the deposited films was controlled by deposition time [1]

3.Results

Figure 2a shows calculated heat volume density $q_V(z)$ at x=0 in 75 nm thick SOQ induced by the 561 nm (black curve) and 364 nm (red curve) light absorption and the temperature field $T(z)$ (blue curve) for the 561 nm light with P = 1 mW. As temperature field $T(x)$ at z=0 (Fig. 2b shows, the heat travels a long distance in the x-direction before being absorbed by the substrate.

Figure 3 shows the power dependencies of the laser-induced heating and their linear fittings for 75 nm thick SOQ and SOS. From the slopes of the dependencies, we find $\Delta T/P$ ~ 6.7 (K/mW) for SOQ and $\Delta T/P$ ~ 1.9 (K/mW) for SOS. Determination of the corresponding k_1 values is graphically shown in Fig. 4. Straight horizontal lines represent experimental dimensionless $P/\Delta T/\lambda/k_2$ values for the 75 nm thick SOQ and SOS while curves show $P/\Delta T/\lambda/k_2$ values calculated for a wide range of k_1/k_2 ratios. The k_1 can be found from the intersection between theoretical curve and corresponding horizontal line. The k_1 ~ 65.5 W/m/K obtained for SOQ appears to be in good agreement with literature while that of SOS ~ 22 W/m/K does not.

Figure 5 shows the theoretical and experimental Si NF thickness dependencies of k: The theoretical calculations were made by assuming in-plane DPT and frequency-dependent boundary scattering [4] Our data (open circles) for SOQ with $h \geq 60$ nm appeared to be in good agreement with theoretical and experimental in-plane Si NF k values from Ref. [4]. However, for SOQ with h < 60 nm, k_1 rapidly drops with a decrease in h, probably, indicating significance of the cross-plane ballistic phonon transport. In this case like in case of SOS, our theoretical model needs modification.

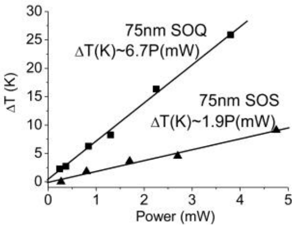

Fig. 3. 75 nm thick SOQ and SOS ΔT (P) dependencies and thei linear fittings.

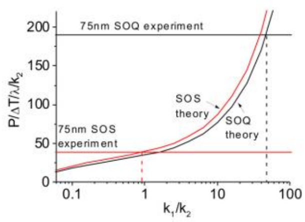

Fig. 4. Calculated dimensionless $P/\Delta T/\lambda/k_2$ parameters vs. k_1/k_2 for 75 nm thick SOQ (black curve) and SOS (red curve). Horizontal lines represent experimental $P/\Delta T/\lambda/k_2$ values.Vertical dashed lines show the determined k_1/k_2 for both cases.

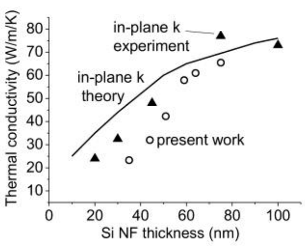

Fig. 5. Thickness dependence of Si NF thermal conductivity k: 1) theoretical and experimental results for in-plane k [4] (black curve and triangles, respectively), and 2) results obtained in this work (open circles).

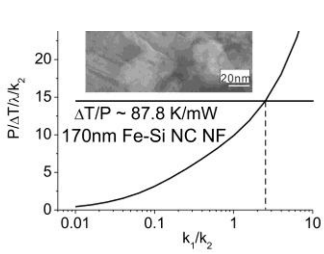

Fig. 6. Dependence of calculated (curve) dimensionless parameter $P/\Delta T/\lambda/k_2$ on k_1/k_2 for 170 nm thick Fe-Si NC NF and experimentally determined value of the parameter (horizontal line). Vertical dashed line shows the determined k_1/k_2. Inset shows TEM image of Fe-Si NC NF film cross-section.

Now let us consider the case with $h > \Lambda$ and k_1/k_2 ~ 1. Figure 6 shows the k_1 determination for 170 nm thick Fe-Si NC NF. The inset shows a TEM image of Fe-Si NC NF cross-section with few tens nanometer grain sizes. The Λ in NC NF is even lower than the grain size. The NF displays very high $\Delta T/P$ ~ 87.8 K/mW and exhibits low k_1 ~ 3.5 W/m/K in agreement with the value obtained using the thermal reflectance method. Despite k_1/k_2 ~ 2.5, the solution is stable (Fig. 6) due to sufficiently large h.

4. Conclusions

We established a Raman spectroscopic k measurement method for absorbing NFs on bulk transparent substrates and applied it for Si and $FeSi_{20}$-2P NFs on SiO_2..

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