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A simple efficient method of nanofilm-on-bulk-substrate thermal conductivity measurement using Raman thermometry

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ABSTRACT

In contrast to known Raman-thermometric measurements of thermal conductivity (k) of suspended Si nano-membranes, here we apply Raman thermometry for k measurement of mono- and nano-crystalline Si films on quartz, which is important for applications in thermoelectricity and nanoelectronics. Experimentally, we measure linear dependence of the laser-induced Raman band downshift, which is proportional to the moderate heating ΔT, on the laser power P. Then we convert the downshift to ΔT and determine the ratio $\Delta T/P$. The actual power absorbed by the film is calculated theoretically and controlled experimentally by the reflection/transmission measurement. Then we calculate $\Delta T_{calc}/P$ for arbitrary film k assuming diffusive phonon transport (DPT). Film k is determined from the condition $\Delta T/P = \Delta T_{calc}/P$. We show that this method works well for films with thickness $h > \Lambda$, where Λ is phonon-mean-free path, even for low-k films like nano-crystalline Si and SiGe. For $h < \Lambda$, despite ballistic phonon transport contribution, this approach works when the in-plane DPT dominates, e.g. in Si films on quartz with $h \geq 60$ nm. We also show that the influence of thermal boundary resistance on the determined k is negligible at this condition. The proposed method is simple and time efficient, as dozen of films can be examined in one hour.

1. Introduction

Measurement of thermal conductivity (k) of nanofilms (NFs) and, especially, Si-based NFs is important for applications in thermoelectricity and nanoelectronics. NF k is reduced compared to bulk material due to phonon boundary scattering and some other effects. Reduced k limits the ability to remove waste heat in microprocessors, light-emitting diodes, memory and solar-cell devices. On the other hand, a reduced k can play a positive role in thermoelectric devices. The efficiency of such devices depends on the thermoelectric figure of merit $ZT = S^2T\sigma/k$, where S, σ and T are the Seebeck coefficient, electrical conductivity, and absolute temperature of material, respectively. Bulk Si was never used in thermoelectric applications due to its high $k \sim 150$ W/m/K and, therefore, low ZT < 0.01 at room temperature. Although Si is a poor thermoelectric material, a reduction of k in Si NFs and nanowires (NWs) can considerably improve their performance [1–9]. Further k reduction can be achieved in nanocrystalline Si-based NFs [10,11]. In the composites consisting 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 [11].

Among a variety of methods of k measurements of Si-based nanostructures, non-contact Raman thermometry is, especially, attractive. At moderate laser-induced heating ΔT, Si optical phonon ~520.5 cm^{-1} Raman band displays downshift $\Delta\omega$ (cm^{-1}) = −0.022ΔT (K) [12]. For unsupported Si particles, overheating can be substantial, i.e. $\Delta T > 1000$ K is possible at rather low laser power $P \sim 1$ mW since there is no efficient heat drain [13]. Contrary, in bulk materials and suspended or supported nanostructures, there is a heat drain. If we correctly take the drain into account then we can extract k from the Raman thermometric data.

Recently, Raman thermometry was utilized to measure k in bulk absorbing materials [14,15], suspended nano-membranes [14,16–22] (NMs) and NWs [23–26]. Even time-domain differential Raman thermometry was developed in recent experiments [27]. The ΔT of the illuminated NM/NW area depends on the k,

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absorption of NM/NW, P, laser beam diameter and the distance of the area from the heat drain. Since the k is the only unknown parameter, one can find it. For ultrathin suspended NMs, ambient environment detrimentally affect ΔT, which is a disadvantage of the Raman thermometry in this case as was shown for grapheme [18].

In contrast to suspended NMs and NWs, here we consider NFs on bulk substrates, in which the effect of ambient environment is much less than that for suspended NMs/NWs. The dominant heat drain is a few millimeter thick substrate supporting NF. Therefore, the experimental Raman measurement procedure is very simple, just standard micro-Raman spectra acquisition. However, the k extraction from ΔT is rather complicated in this case. We solve this problem for absorbing NF on bulk non-absorbing low-k substrate and measure k of Si NFs and composite NC NFs of Si and Fe silicide (Fe-Si NC) NFs on quartz. Fe-Si NC is similar to Ni-Si NC but more advantageous for industrial fabrication due to its lower cost. Additionally, we fabricated NC SiGe film and showed further decrease in the thermal conductivity due to the enhancement of the alloy phonon scattering. Our presented method for k measurement for NF-on-substrate is simpler and more efficient than alternative methods. A dozen of NFs can be examined in one hour.

2. Theoretical calculation of the laser-induced heating of absorbing nanofilm on transparent bulk substrate

As shown in Fig. 1a, we consider an absorbing layer 1 with complex refractive index $N_1 = \sqrt{\varepsilon_1} = n_1 + i\kappa_1$ and thickness h ($0 \le z \le h$) on a semi-infinite substrate (layer 2) with the refractive index $N_2 = \sqrt{\varepsilon_2} = n_2$ ($z > h$). Laser light propagates from a semi-infinite media with the refractive index $N_0 = \sqrt{\varepsilon_0} = n_0$ ($z < 0$), hits the absorbing layer 1 and propagates further into the layer 2. The laser-light-power-flux density is:

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

where r_0 is the radius of the focused laser beam (Fig. 1a, b). The volume density of heat per second generated by the light absorption can be expressed as follows [28]:

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

where ω is the light frequency, ε_1'' is the imaginary part of dielectric constant and E_1 is the electric field of light in the layer 1. We calculate E_0, E_1 and E_2 using exact analytical solution of Maxwell equations for layered medium taking into account reflection, absorption and interference [29]. Fig. 2a shows functions $q_V(z)$ for 75 nm thick Si on quartz (SOQ) at the center of the laser beam ($x = 0$, $r_0 = 250$ nm) for the wavelength $\lambda = 561$ nm ($n_1 = 4.05, \kappa_1 = 0.035$) and for $\lambda = 364$ nm ($n_1 = 6.45, \kappa_1 = 2.76$). The 364 nm light is nearly completely absorbed in ~20 nm thick layer with no noticeable interference while the 561 nm light displays interference. At $\lambda = 561$ nm, the SOQ absorption A is ~7.4% with reflection $R \sim 16.1\%$ and transmission $Tr \sim 76.5\%$.

For experimental control of the film A, we measured reflection/transmission spectra. Then we determined A from $A = 100\% - R - Tr$. Fig. 3 shows experimental and theoretical reflection spectra of 59 nm and 64 nm thick SOQ. The agreement between experiment and theory is very good. Statistically, we estimated that the

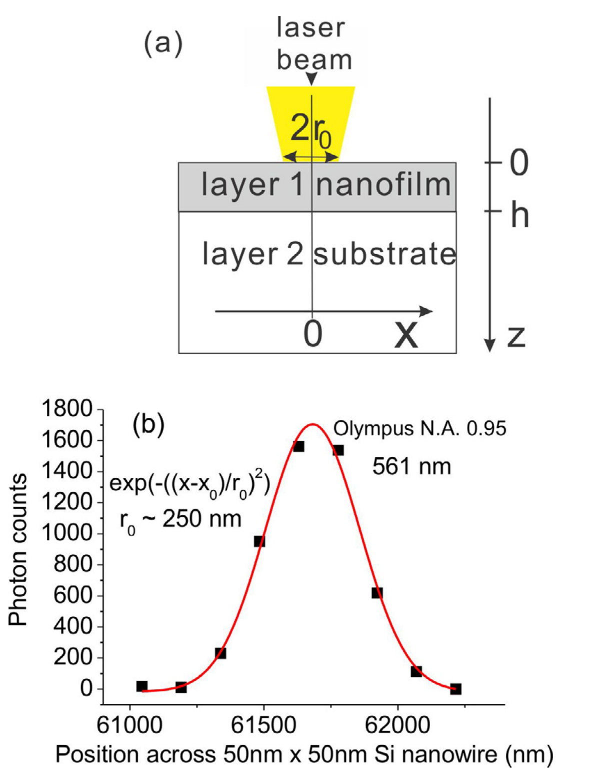

Fig. 1. (a) Schematic view of laser illumination of absorbing NF on transparent substrate; (b) light intensity distribution in the focused 561 nm laser beam measured while scanning the beam across 50 nm × 50 nm Si NW and detecting NW Raman signal (black squares). Red curve shows Gaussian fitting of the experimental points. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

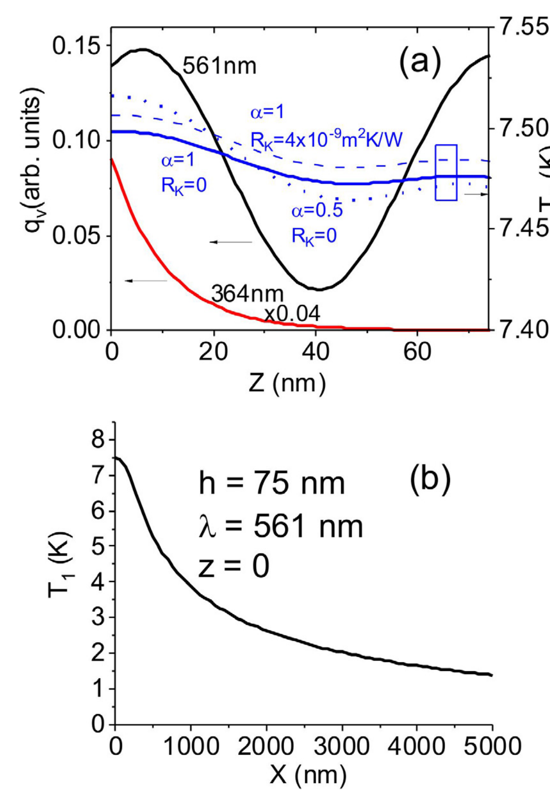

Fig. 2. (a) The volume density of heat per second $q_V(z)$ at $x = 0$ generated in 75 nm thick SOQ by the 561 nm (black curve) and 364 nm (red curve) absorbed light (r_0 = 250 nm) correspond to the left axis; the temperature (heating) field $T_1(z)$ for the 561 nm light with P = 1 mW for three cases (blue curves): (1) no anisotropy of the film thermal conductivity $\alpha = 1$ and no Kapitza resistance $R_K = 0$ (solid); 2) $\alpha = 1$ and $R_K = 4 \times 10^{-9}$ m^2 K/W (dashed); (3) $\alpha = 0.5$ and $R_K = 0$ (dotted) correspond to the right axis; (b) temperature (heating) field $T_1(x)$ at $z = 0$ for 75 nm thick SOQ. The $T_1(x)$ curves corresponding to three cases with different α and R_K shown in (a) cannot be distinguished in this scale. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)