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Precise Thermal Characterization of Confined Nanocrystalline Silicon By a 3ω Method

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1. Introduction

Due to a strong quantum confinement effect, the thermal conductivity Λ and heat capacity per unit volume C of a nanocrystalline silicon (nc-Si) layer prepared by electrochemical anodization are extremely lowered in comparison to those of single crystal silicon (c-Si). Based on this big difference in the thermal properties between nc-Si and c-Si, we have developed thermally induced ultrasound emitters as shown in **Fig. 1** [1-4]. This device has many advantages over conventional airborne ultrasound devices such as piezoceramic transducers: the ultrasound emission is obtained without any mechanical vibrations and exhibits a flat frequency response in a wide range.

For designing the device, Λ and C are key parameters, since the acoustic output is inversely proportional to the square root of the product ΛC. In this article, we present a precise measurement of Λ and C using a dynamic approach so called the 3ω method [5-9]. To our knowledge, no papers devoted to the heat capacity measurements in nc-Si layers have yet been published.

2. Experimental

An nc-Si layer was formed by electrochemical anodization of (100) p^--type c-Si wafer. After the anodization in an ethanoic HF solution at a current density of 20 mA/cm^2 for 20 min, the thickness d and porosity p of the anodized nc-Si layer was 30 μm and 55%, respectively. According to the results of photo-acoustic and photoluminescence measurements, the band gap and emission band of the sample were ~2.5 eV and yellow, respectively.

Figure 2 illustrates the experimental setup for the 3ω method employed here. A thin metal line was formed on the nc-Si layer by vacuum evaporation and subsequent photolithography, and used as both a heater and thermometer. The length, width, and resistance of the electrode are denoted as l (e.g. 3 mm), $2b$ (e.g. 25 μm), and R_0, respectively. The metal line was heated by an oscillating current I_0 with frequency ω using a signal generator. The produced Joule's heating causes a temperature oscillation $\Delta T(\omega)$ at the surface. Using a bridge circuit and a lock-in amplifier, $\Delta T(\omega)$ were detected by small voltage oscillations at third harmonic component $V_{3\omega}$ caused by the temperature-dependent resistance of the metal line. Here $\Delta T(\omega)$ was determined using $2V_{3\omega}/(\alpha I_0 R_0)$, where α is the temperature coefficient of the resistance ($=1/R_0 \cdot dR_0/dT$) of the metal line. The measurements were performed in a vacuum and at room temperature.

The theoretical $\Delta T(\omega)$ is given by the differential equation of heat conduction in the solid state [6,7]. At low frequencies (<1 kHz) in which the thermal diffusion length is much longer than d, $\Delta T(\omega)$ is a summation of two components: one from the substrate, $\Delta T_s(\omega)$, and the other from the nc-Si layer, $\Delta T_n(\omega)$. The quantities $\Delta T_s(\omega)$ and $\Delta T_n(\omega)$ are given approximated by Eq. (1) and Eq. (2) (see Appendix). The quantity of $\Delta T_s(\omega)$ is obtained from the well-known thermal parameters of c-Si [10]. Since the difference between the experimental $\Delta T(\omega)$ and $\Delta T_s(\omega)$ corresponds to the nc-Si component $\Delta T_n(\omega)$, we can determine the Λ value from Eq. (2). At high frequencies, in contrast, the substrate component ΔT_s is negligible, and then $\Delta T(\omega)$ is given by Eq. (3) (see Appendix). Using the Λ value determined from the low-frequency data mentioned above, the C value can be obtained from Eq. (3).

3. Results and Discussion

Figure 3 shows the measured frequency dependence of the temperature oscillations. According to the data lower than 200 Hz, the Λ value is 1.22 W/(m·K) in good agreement with that measured by Drost *et al.* [11]. The solid line is a fitting one to Eq. (1) and Eq. (2). As the frequency increases beyond ~ 10^3 Hz, $\Delta T(\omega)$ decreases rapidly. In the high frequency region, the slope of $\Delta T(\omega)$ vs. $\ln(\omega)$ characteristic (represented by dashed line) suggests that $\Lambda \sim 1.2$ W/(m·K). This agrees with that determined from the low-frequency data. The corresponding C value is ~ 0.15×10^6 $J/(K \cdot m^3)$. The heat capacity is considerably lower than the value simply estimated from the porosity of the sample. The observed marked decline in the heat capacity is another indication of the nature of nc-Si as a confined material with a significant band gap widening and complete carrier depletion.

4. Summary

Based on a 3ω method, we have determined the thermal conductivity and heat capacity of nc-Si layer. As summarized in Table I, the thermal properties of nc-Si show very unique features in comparison to those of c-Si, quartz glass, and thin SiO_2 film. This result provides useful information for development of an integrated efficient ultrasound emitter and possibly of novel functional acoustic devices.

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Appendix

$$\Delta T_s(\omega) = \frac{P}{\pi\Lambda_s}\left\{\frac{1}{2}\ln\left(\frac{\Lambda_s}{C_s b^2}\right) + 0.932 - \frac{1}{2}\ln(2\omega)\right\} \quad (1)$$

$$\Delta T_n(\omega) = \frac{Pd}{2b\Lambda}, \quad (2)$$

where P, Λ_s, and C_s are the power dissipation per unit length of the metal line, the thermal conductivity of c-Si, and the heat capacity per unit volume of c-Si, respectively.

$$\Delta T(\omega) = \frac{P}{\pi\Lambda}\left\{\frac{1}{2}\ln\left(\frac{\Lambda}{Cb^2}\right) + 0.932 - \frac{1}{2}\ln(2\omega)\right\}. \quad (3)$$

Table I. Thermal parameters of nc-Si, c-Si, quartz glass, and thin SiO_2 film.

Material	Thermal conductivity (W/(m•K))	Heat capacity per nit volume (MJ/(K•m^3))	Ref.
nc-Si	1.2	0.15	This work
c-Si	148	1.67	[10]
Quartz glass	59.9	1.61	[10]
Thin SiO_2 film	0.9~1.4	1.6~1.9	[9]

Fig. 1. Photograph of the fabricated nc-Si ultrasound device. The chip (3 × 3 mm) is mounted on a TO-5 housing.

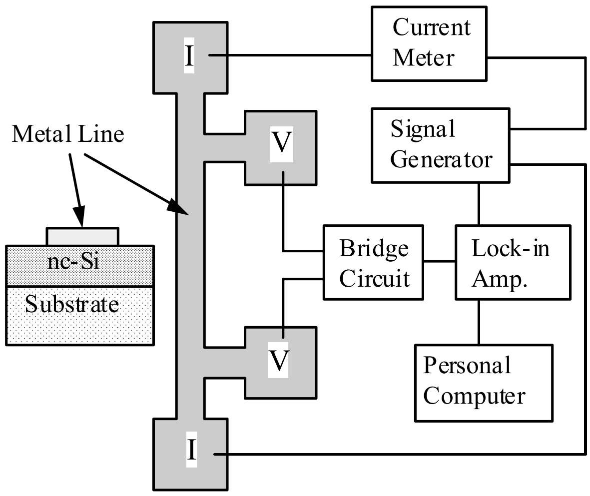

Fig. 2. Schematic sample structure and experimental configuration. Details of the 3ω method are available in the literature (ref. 6-10).

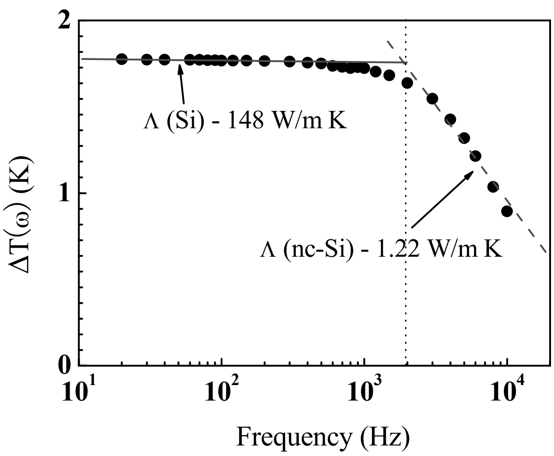

Fig. 3. Frequency dependence of the measured temperature oscillation at the surface of the nc-Si sample (closed circles). The fitting lines in the low and high frequency regions are also shown.