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Interfacial thermal resistance between porous layers: Impact on thermal conductivity of a multilayered porous structure

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ABSTRACT

Features of thermal transport in multilayered porous silicon nanostructures are considered. Such nanostructures were fabricated by electrochemical etching of monocrystalline Si substrates by applying periodically changed current density. Hereby, the multilayered structures with specific phononic properties were formed. Photoacoustic (PA) technique in gas-microphone configuration was applied for thermal conductivity evaluation. Experimental amplitude-frequency dependencies were adjusted by temperature distribution simulation with thermal conductivity of the multilayered porous structure as a fitting parameter. The experimentally determined values of thermal conductivity were found to be significantly lower than theoretically calculated ones. Such difference was associated with the presence of thermal resistance at the interfaces between porous layers with different porosities arising because of elastic parameters mismatch (acoustical mismatch). Accordingly, the magnitude of this interfacial thermal resistance was experimentally evaluated for the first time. Furthermore, crucial impact of the resistance on thermal transport perturbation in a multilayered porous silicon structure was revealed.

1. Introduction

For decades, the drive for faster, cheaper computing and its associated device miniaturization have served to push scientists and engineers to develop materials, tools, processes, and design methodologies. State of the art electronic devices already operates with critical dimensions in the tens of nanometers. Development of the next generations of integrated circuits (ICs), three-dimensional (3D) integration and ultra-fast high-power density transistors has led to a steep increase in microprocessor chip heat flux and growing concern over the emergence of on-chip hot spots. Understanding thermal transport at the nanoscale is therefore crucial for a fundamental description of energy flow in nanomaterials, as well as a critical issue toward achieving optimal performance and reliability of modern electronic, optoelectronic, photonic devices and systems. Thermal transport at the nanoscale is fundamentally different from that at the macroscale and is determined by the distribution of carrier mean free paths and energy dispersion in a material, the dimension of the structure and the distance over which heat is propagated. The opportunity to shape new nanostructures that efficiently scatter phonons, reducing the thermal conductivity, without altering the electrical properties of the material enables the potential implementation of proficient thermoelectric devices to work as coolers or power generators from waste heat [1–4]. Recently, there has been much interest in the thermal conductivity of semiconductor superlattices (SLs) due to their promising applications in a variety of devices.

Since important surface-to-volume fraction in nanostructured materials, heat conduction across solid-solid interface predominates thermal transport there. Particularly, in SLs, multiple interfaces between different materials play a critical role in the thermal conductivity reduction [5,6]. Cutting-edge experimental techniques have enabled the measurements of the in-plane [7] and cross-plane [8] thermal conductivity in SLs. Experiments revealed that the thermal conductivity of SLs is strongly anisotropic and significantly lower than that estimated from the bulk value of the constituent materials and even smaller than the thermal conductivity of the equivalent composition alloys [9–12]. Theoretical investigation has established that the diffuse interface scattering induces the decrease of the in-plane (and, in part, the cross-plane) thermal conductivity, while the thermal boundary resistance (TBR) between adjacent layers is a key factor in the cross-plane thermal-conductivity reduction [13,14].

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It is well-known, that stable mesoporous silicon thin films have thermal conductivity values two orders of magnitude lower than the thermal conductivity of bulk crystalline silicon [15,16]. Three orders of magnitude reduction have recently been achieved by applying additional treatment [17]. Thus, porous silicon is a promising candidate as a heat insulating material for micro- and nanoelectronic devices, especially taking into account its full compatibility with CMOS processing. Accordingly, a systematical study of the heat transport in various porous silicon systems can give physical insight for the further improvement of thermal engineering in silicon-based elements.

In this article, we propose to study the thermal properties of porous silicon based multilayered nanostructures. The aim of our work is to investigate thermal resistances of the interfaces between layers of different porosities. Photoacoustic (PA) technique in a classical configuration was used to probe the effective room temperature thermal conductivity across the multilayered porous nanostructures. Experimental analysis of thermal transport in the considered structure allows us to estimate interfacial thermal resistance values for the different ratio of porosity between successive layers.

2. Methods

2.1. Experimental details

Porous silicon multilayer structures were prepared by electrochemical anodization of (100)-oriented p^+- type boron-doped (0.01–0.02 Ohm·cm) monocrystalline silicon (c-Si) wafer (initial wafer thickness l_{Si} = 510 μm), which were previously immersed in a hydrofluoric acid (HF) solution to remove any native oxide. The electrochemical etching was carried out in an electrolyte based on 49% HF and pure ethanol (at a volumetric ratio of 1:2). Anodization current density sequences (programmed by a Keithley DC power supply) corresponding to values in the range 7.5–80 mA/cm^2 was applied (see Table 1 in Appendix). The etched area was 1.0 cm^2, the total number of porous bilayers was 30. Porosity values controlled by the etching conditions have been extracted from preliminarily established calibration curves. Thicknesses of the etched porous layers have been measured from the scanning electron microscopy (SEM) images obtained by a MIRA 3 Tescan microscope. The cross-sectional SEM images of samples 1 and 2 are shown, for example, in Fig. 1.

Investigation of thermal transport properties was performed by applying the PA technique in classical configuration [17,18], at room temperature. All samples placed in the PA cell were irradiated by a laser diode module with an output optical power of about 60 mW (see Fig. 2) and a λ = 405 nm spectral wavelength. The light with an electrically modulated intensity was uniformly distributed over the whole sample surface by an optical system. The PA signal was detected by a microphone coupled to a lock-in nanovoltmeter in the 15–1000 Hz frequency range and compared to the reference signal delivered by the current generator.

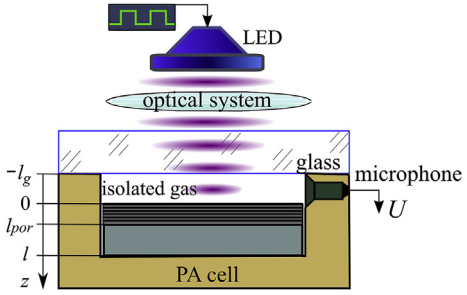

Fig. 2. Experimental PA set-up in classic configuration.

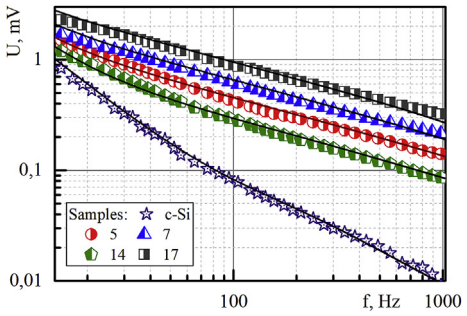

Fig. 3. Experimental amplitude-frequency dependencies of the PA signal for samples 5, 7, 14, 17, and the reference monocrystalline Si. The solid lines correspond to the calculated AFC obtained from the temperature distribution simulations described below.

As an example, experimental amplitude-frequency characteristics (AFC) of porous samples 5, 7, 14, 17, and the reference monocrystalline Si are presented in Fig. 3.

2.2. Simulation details

Since the exciting UV light is absorbed within the first porous monolayer, its absorption coefficient (α) can be calculated from the Bruggeman's model [19,20] based on an effective medium approximation. Therefore, the permittivity of the porous monolayer ε_{por} can be calculated as a function of porosity (ξ) with the following equation:

$$\xi\frac{1-\varepsilon_{por}}{1+2\varepsilon_{por}} + (1-\xi)\frac{\varepsilon_{Si}-\varepsilon_{por}}{\varepsilon_{Si}+2\varepsilon_{por}} = 0 \quad (1)$$

where ε_{por} and ε_{Si} are the complex permittivity of porous and bulk silicon, respectively. The well-known complex permittivity value of the bulk monocrystalline silicon can be found in the literature [21,22]. Absorption coefficient α_{por} of the porous layer can be deduced as follow (see Table 2 in Appendix):

$$\alpha = 4\pi k/\lambda \quad (2)$$

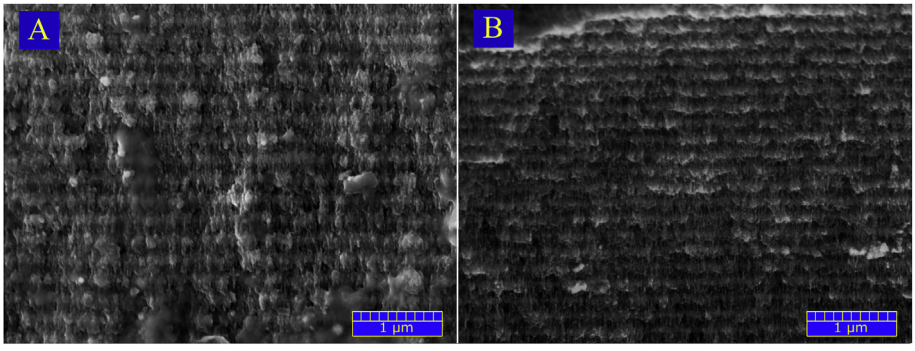

Fig. 1. Cross-sectional SEM images of porous silicon multilayers of samples 1 (A) and 2 (B).