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Thermal boundary resistance correlated with strain energy in individual Si film-wafer twist boundaries

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ABSTRACT

Nanoscale interfaces, such as grain boundaries (GBs) within a polycrystalline material, play an important role in suppressing the phonon heat transport. The interfacial thermal resistance R_K of a GB has a strong dependence on the detailed interfacial atomic structure, including the misorientation between two grains and GB dislocations. Along this line, numerous molecular dynamics simulations on R_K have been carried out on a twist Si GB. Owing to the challenge of measuring such a GB within a bulk material, these simulations are rarely compared with experimental data. In this work, a super-flexible 70-nm-thick Si thin film was hot pressed onto a Si wafer to represent a twist GB. The R_K of the film-wafer interface was measured as a function of the rotation angle between the film and the wafer. The experimental data were further compared with an analytical model to interpret the twist angle dependence of the measured R_K. It was found that the strain part of the grain-boundary energy is correlated with the measured twist-angle-dependent R_K.

1. Introduction

A grain boundary (GB) inside polycrystalline materials can transmit or reflect incident phonons. Such interfacial phonon scattering introduces an interfacial thermal resistance R_K, known as the Kapitza resistance [1,2]. With a high volumetric density of GBs, phonon transport within a material with fine grains can be largely suppressed by GBs [3]. The reduction of lattice thermal conductivity k_L by GBs has been exploited to develop better thermal insulation and thermoelectric materials [4]. In addition, the R_K for GBs and other interfaces can significantly impede heat spreading in electronic devices, thus creating challenges for thermal management [3,4].

In a polycrystal, various GBs with different structure and energies are present simultaneously, which can be described through the GB character distribution [5]. GBs within this distribution have varying crystal misorientations (with five macroscopic degrees of freedom) and varying atomic structures at the interface (additional microscopic and atomic degrees of freedom). The total GB energy (γ_{GB}) is given as $\gamma_{GB} = \gamma_{core} + \gamma_{strain}$, where γ_{core} and γ_{strain} are the energy due to broken bonds across the interface and the energy due to GB strain fields, respectively. The atomic structure of many GBs in their low total energy atomic configuration minimizes the number of broken bonds across the interface and is viewed as arrays of GB dislocations separated by strained perfect crystal [5]. In contrast, GBs described as disordered or amorphous tend to contain larger amounts of broken bonds across the interface and, while they usually have low amounts of strain energy, they tend to be higher in total energy [6]. It is widely accepted that the structure of a GB can influence phonon transport across it [3,4]. However, detailed studies are required to determine which attributes of this GB structure and energy are the most important for phonon transport.

In some molecular dynamics (MD) studies, the total GB energy (γ_{GB}) is proposed to be associated with R_K [7–9]. Additionally, the local strain field that forms at a GB has been shown to affect the phonon transport [10]. In the widely used acoustic mismatch model (AMM) and diffuse mismatch model (DMM), however, the complexity associated with the structure and energy of a GB is usually neglected. Assuming specular phonon scattering on an interface, the AMM fails to predict the GB R_K because of the same acoustic impedance between two grains made of the same material [3]. Developed for a rough interface with diffusive phonon scattering, the DMM [2] instead predicts a fixed GB phonon transmissivity τ_{GB} of 0.5 at all frequencies due to structure symmetry of a GB [11]. Because high-frequency phonons are anticipated to be

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scattered stronger by a GB, τ_{GB} should decrease with an increased phonon frequency, and the DMM is thus expected to be over-simplified [8,12,13]. More advanced MD simulations have been carried out to predict the R_K for various GBs [8,14–17]. However, experimental measurements are often lacking to validate these predictions, mainly due to the challenge in measuring a single GB within a three-dimensional bulk material [4]. In most data analysis, the averaged R_K of all GBs within a polycrystalline material can be extracted using an effective medium formulation, whereas the difference between varied GBs is neglected [18–22].

Due to the challenge in measuring a single GB within a bulk material, bonding between identical wafers with relative rotation has been used to gain insights into the phonon transport across twisted GBs [23,24]. In these studies, two identical crystals are rotated relatively around an axis that is perpendicular to the GB plane. A phonon focusing image has also been taken for twist-bonded wafers [25]. A major challenge for such studies is maintaining high-quality bonding across the whole wafer. This requires both wafers to be atomically flat to avoid the creation of voids or unintended structural defects. From this perspective, much better thermal bonding can be achieved using super-flexible thin films as one of the crystals. This is essentially due to a large initial film-wafer contact area and negligible separation or fracture during the bonding process. Even for a film and a wafer with a large lattice mismatch and/or a large twist angle, a high-quality interface can still be obtained, which is challenging when bonding two rigid wafers. For device fabrication, extremely high-quality bonding has been demonstrated between a 200-nm-thick and millimeter-sized Si membrane and a Ge wafer [26]. High-resolution transmission electron microscopy (TEM) shows a thin ~1 nm region for the bonded Si-Ge interface. With moderate pressure at 473 K, stacked nanometer thick membranes can be used to form a Si-Ge- superlattice with a cross-plane k_L below 2 W/m·K [27]. For phonon transport studies, these high-quality interfaces for film-wafer or film-film bonding provide ideal model systems for real GBs that are formed by hot pressing nanopowder into a bulk material [28,29].

In this work, thermal investigation of twist GBs was performed with a 70-nm-thick (100) Si thin film hot pressed onto a (100) Si wafer. The high-quality interfaces achieved via film-wafer bonding was confirmed by TEM studies and enabled thermal studies of GBs across a large range of misorientation angles. Temperature-dependent interfacial R_K between the film and the wafer was carried out for different twist angles between the film and the wafer. GBs with a low (<15°) and high twist angle were measured, which was enabled by the film-wafer bonding. This broad range of twist angles reached beyond previous studies, which used wafer-wafer bonding methods and were constrained to small twist angles [23,24]. Detailed electron microscopy studies were carried out on representative samples to reveal the interfacial atomic structures. An analytical model based on such an interfacial structure was used to interpret the observed twist-angle dependence of R_K. Although some earlier studies associated R_K with the total GB energy γ_{GB} [7–9], this work suggests a correlation between R_K and the strain-part of the GB energy, γ_{st}. Our results provide new insights into phonon transport across GBs, which are important for technologies ranging from microelectronics to energy materials.

2. Material and methods

2.1. Sample preparation

The (100) thin films used were released from a commercial silicon-on-insulator (SOI) wafer and transferred onto another (100) Si wafer with 500 μm thickness. The film transfer was performed in diluted hydrogen fluoride (HF) acid to prevent oxidation of Si, which was found on the bonded interface of Si wafers [24] and at GBs within hot-pressed nanocrystalline silicon [12]. The whole transfer process followed that used for graphene, with a photoresist layer to protect the film during the transfer process [30]. This photoresist layer was later dissolved in acetone. Before the hot press, the film was further annealed at 473 K to ensure good sample-substrate contact. A thin SiO_2 layer with 20 nm thickness was then deposited onto the sample to avoid possible contamination during the following hot press. The hot press was then carried out under N_2 protection at around 1223 K with 1-min holding. The rapid heating was provided by an induction heater, as used for bulk-material synthesis [31]. Roughly 50 MPa pressure was applied with a graphite die and this pressure was uniformly distributed across the film using a soft graphite foil. After the hot press, the SiO_2 layer was etched off by HF. The residual graphite was removed together with the SiO_2 layer, as shown in Fig. 1a.

The employed hot-press condition should be compared with that used for nanocrystalline bulk Si, where a higher hot-press temperature (1327–1523 K) with 0–2.5 min holding was required to obtain up to 99.5% theoretical density [12]. For bulk materials, this higher hot-press temperature is required to soften the irregular-shaped nanopowder for better compaction. For film-wafer bonding, a lower bonding temperature was usually used because the film and wafer were in good contact already. In practice, further increasing the hot-press temperature may cause cracks on the film when the twist angle between the film and the wafer was large. To further improve the bonding quality, all samples were finally sealed in a vacuum tube and annealed at 1173 K for 1 hr. The interfacial R_K for all samples was measured after annealing, and selected samples were also measured right after the hot press.

Cross-sectional TEM studies were also carried out on representative samples with a low to a high film-wafer twist angle (Fig. 1b and c). GB regions with around 3.0 nm and 4.25 nm thicknesses were observed in the TEM images of samples with 3.4° and 86.5° twist angles, respectively. Interfacial layers were observed with no apparent structure (appears amorphous) when viewed in this direction, as suggested by MD simulations previously [32]. Some GB structures were found later within this “amorphous” region [33]. These interfacial layers were thicker than the ~1 nm interfacial layer for the previous bonding between a Si film and a Ge wafer [26], which was performed only at 673 K for 30 min. For all samples, further checking with energy-dispersive X-ray spectroscopy suggested almost uniform surface natural oxidation across the entire cross section, i.e. there is no amorphous SiO_2 layer at the film-wafer interface. In previous thermal studies using wafer-wafer bonding, an extra SiO_2 layer was found on the bonded interface so that the measured R_K did not represent that for a “clean” GB in bulk materials [24]. This oxide layer was prevented in this study with careful oxygen protection during the hot press.

To better understand the GB region, TEM images normal to the GB plane (i.e. plane view) were also taken to reveal that the interfacial region is not amorphous but shows a periodic GB dislocation structure for the sample with a 3.4° twist angle (Fig. 1d and e). This further confirms that clean interfaces with low-energy atomic configurations were synthesized. In the literature, similar TEM studies were carried out for the bonding between the device layer of a SOI wafer and a Si wafer [34]. A similar dislocation network was observed in this earlier study. However, the device layer was not released from the SOI wafer during the thermal bonding, which differs from the current work.

2.2. Offset 3ω measurement

The obtained film-on-substrate structure was measured with an offset 3ω method [35] by comparing the measurements in regions