Thermal Interface Properties of Carbon Nanotubes on Diamond

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This work reports on the experimental study of thermal contact conductance of interfaces that include diamond films. Polycrystalline diamond thin films were directly synthesized on silicon chips using plasma-enhanced chemical vapor deposition. In addition, a hybrid structure consisting of a carbon nanotube-diamond (CNT/diamond) combination was fabricated. Thermal contact conductances of copper-diamond and and copper-CNT/diamond interfaces were experimentally measured with a steady-state contact conductance technique, and relatively high resistances were observed.

A major challenge for achieving thermally reliable semiconductor devices involves the reduction of thermal interfacial and constriction resistances. While the power density of newly developed electronic devices continues to increase, the total thermal budget, i.e. the difference between the junction operating temperature and cooling medium's inlet temperature has remained unchanged. Reduction of thermal contact resistance at solid interfaces enables larger temperature differences between the convective surfaces and cooling medium. On the other hand, the heat generation in most devices is not distributed uniformly; therefore, spreading the heat from the hottest spots effectively and maintaining an acceptable temperature at these spots are also critical to thermal reliability [1-3]. Besides metal-particle-laden phase change materials (PCMs) and thermal gels [4-6], low temperature/self-reactive solders [7] are also being considered as high conductance interface materials to satisfy contemporary heat dissipation requirements. However, issues such as dry-out/pump-out and mechanical fatigue compromise the long-term reliability of many of these materials [5]. Further, for most soft thermal interface materials, high conductance is achieved primarily by enhancing the gap materials' spreadability and elasticity, but adequate heat spreading from hot spots remains a significant issue.

Recently, significant attention has focused on using highly thermal conductive carbon nanotubes (CNTs) for thermal contact conductance enhancement. Among the several reported works, Xu and Fisher [8] have reported the lowest resistance values of 19.8mm^{2}K/W and 5.2mm^{2}K/W under moderate pressures for copper-silicon interfaces with dry CNT arrays and PCM-CNT arrays, respectively. Ngo et al. [9] used copper as a gap filler to enhance the stability and thermal conductance of carbon nanofiber (CNF) arrays. They reported a resistance of 25mm^{2}K/W under a pressure of 60psi for Cu-Si interfaces. However, the CNT/CNF arrays are electrically conductive and therefore may not be suitable for all electronics applications. Meanwhile high-quality PECVD diamond is a good electrical insulator and with high resistance to abrasion [10]. Further, with in-plane and transverse thermal conductivities of 500W/(mK) [11] and more than 1000W/(mK) [12] respectively, diamond films can also be excellent heat spreaders.

In the present work, a combination of polycrystalline diamond films and CNT arrays have been fabricated for possible thermal contact conductance enhancement. Diamond thin films were deposited on bare silicon chip surfaces and then coated with well anchored CNT arrays. Both diamond and CNT layers were directly synthesized by plasma enhanced chemical vapor deposition (PECVD) in the same reactor, and thermal contact resistances have been experimentally measured. The effects of the CNT array thickness and topologic characteristics of the diamond film are discussed.

In the present work, thermal contact resistances of interfaces under different conditions were experimentally measured with a reference calorimeter setup based on axial 1-D steady-state heat conduction [13]. A double side polished silicon wafer was diced into the 10mm×10mm chips, and then the diamond thin films were directly deposited on the both sides of these chips. Each diamond thin film growth process involved two growth phases: nucleation to form the diamond crystal seeds on the bare substrate and continuous film growth. Initially, the polished silicon chips were heated to 700C in a 200 sccm hydrogen flow with a chamber pressure of 15 torr. After a 400W microwave plasma was excited and a 250Vdc substrate bias was applied, 10 sccm methane was introduced into the PECVD chamber to begin the 1-hour nucleation. After nucleation, the substrate bias was disengaged, and the chamber pressure, methane flow rate and plasma power were adjusted to be 40 torr, 2 sccm and 1000W respectively for the 7-hour thin film growth. The typical morphology and structure of the diamond films synthesized in the present work are depicted in Fig. 1. The images of tilted plan views (film surface, Fig. 1(a), and substrate edge, Fig. 1(b)) and a section view (Fig. 1(c)), illustrate that the substrate surface is fully covered a continuous polycrystalline diamond film of thickness ~4μm, and the diamond grain sizes range from 0.5 to 4μm. Figure 1(c) also shows that the deposited film exhibits typical columnar grain growth. Further, a section view with higher magnification (Fig. 1(d)) illustrates that diamond film consists of two distinct layers. The 400nm-thick layer of small crystal grains was deposited during the nucleation growth. On this seeding layer, a larger-grain diamond film was synthesized with a columnar crystal habit.

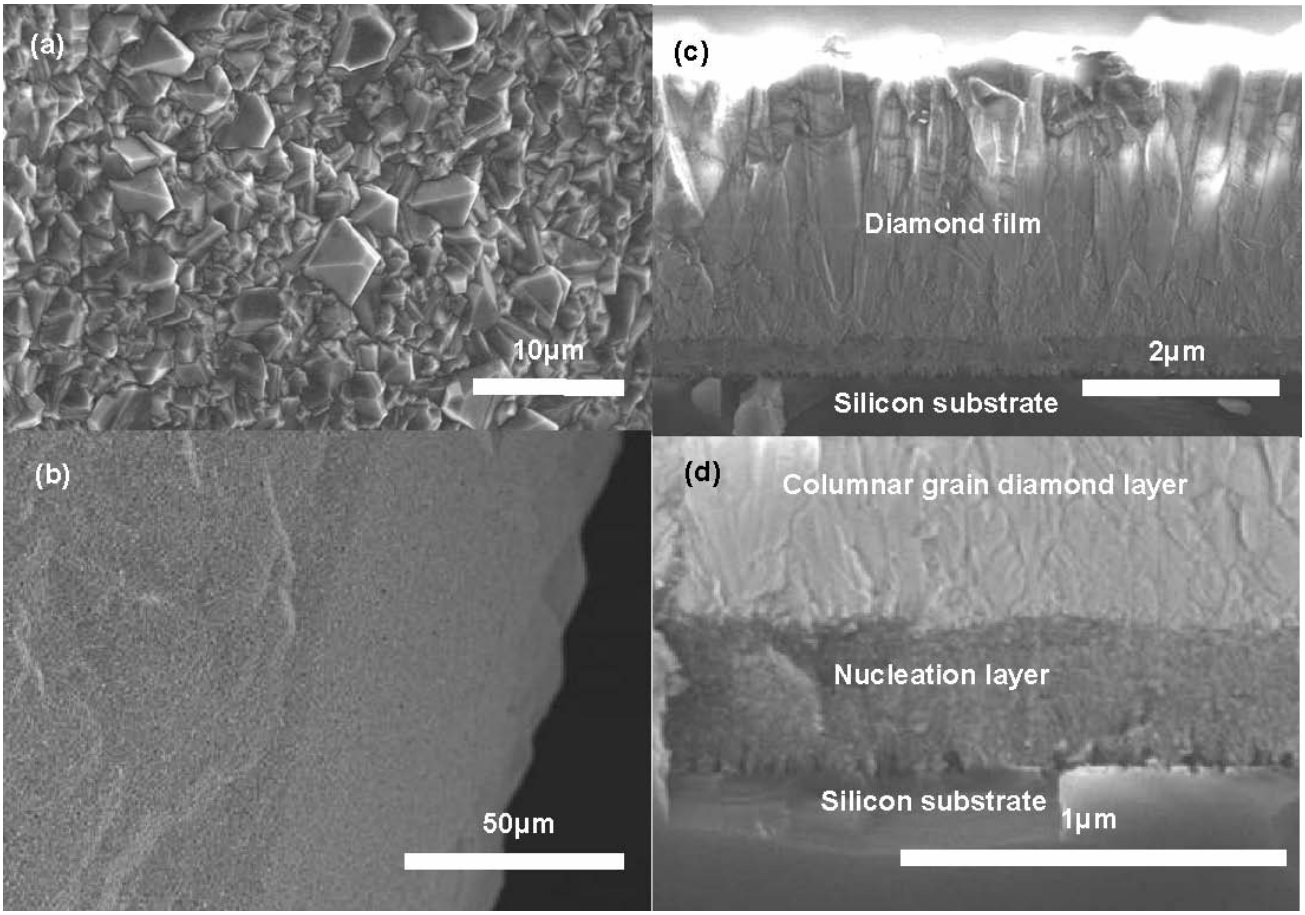

Figure 1. SEM images of a polycrystalline diamond thin film deposited on silicon chip surface with PECVD (a), a tilted view of the diamond film surface, showing the grain sizes ranging from 0.5 to 4 µm (b), the edge of the silicon chip, showing the substrate was fully covered with diamond film (c), a section view showing the structure of a diamond film on silicon chip, the film thick is approximately 4 µm (d), a section view with high magnification showing the nucleation layer.

In prior work, we have developed a tri-layer catalyst configuration [8] that produces well anchored and vertically oriented CNT arrays on metal and non-metal substrates. Figure 2 contains an SEM image of a hybrid structure that consists of a 3µm thick diamond film and a well anchored and vertically oriented CNT array of 8µm height. The CNT array was deposited with a 20-minute growth time under typical conditions [8], *i.e.* chamber pressure of 10 torr, susceptor temperature of 800C, microwave power of 150W, and H_2 and CH_4 mass flow rates of 72 and 8 sccm, respectively. The

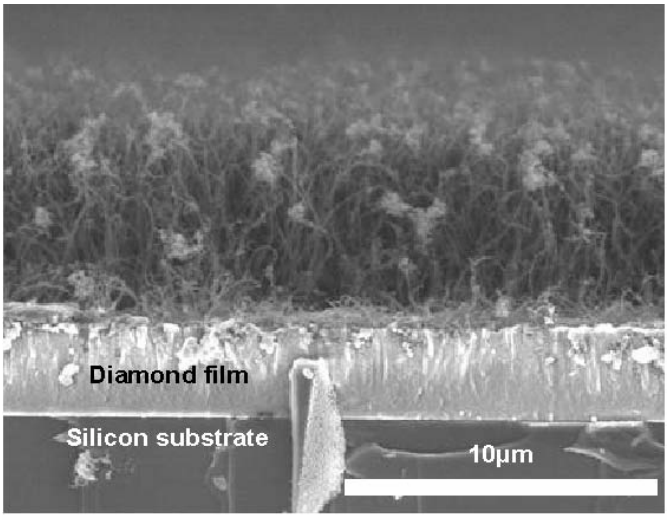

Figure 2. A section view of the CNT-diamond film on a silicon substrate.

density of the CNT array was estimated to be approximately 200 million CNTs per square millimeter, and the CNT diameters ranged from 15 to 50 nm.

In the present study, thermal contact resistances for Cu-diamond and Cu-CNT/diamond interfaces were measured with double-side diamond and double-side CNT-diamond composite specimens that were fabricated in above-mentioned processes, and the results are presented in Fig. 3. In each test, new copper reference bars were used. The thermal contact resistance curve for a Cu-Cu interface is also included in the figure for reference.

The measured bulk thermal contact resistances for the Cu-diamond interface ranged from 5.27 to 9.74 cm^2K/W and were approximately 4 cm^2K/W higher than those of the Cu-Cu interface. The CNT-diamond composite exhibited reduced contact resistances as compared to the diamond-copper interface with a resistance range from 3.16 to 7.64 cm^2K/W. But in contrast to the anticipation of that the 8μm-height CNT array on the rough diamond film surface would bridge most gaps between the copper and diamond surfaces, the diamond-CNT composite on the present specimens did not produce the expected contact conductance enhancement.

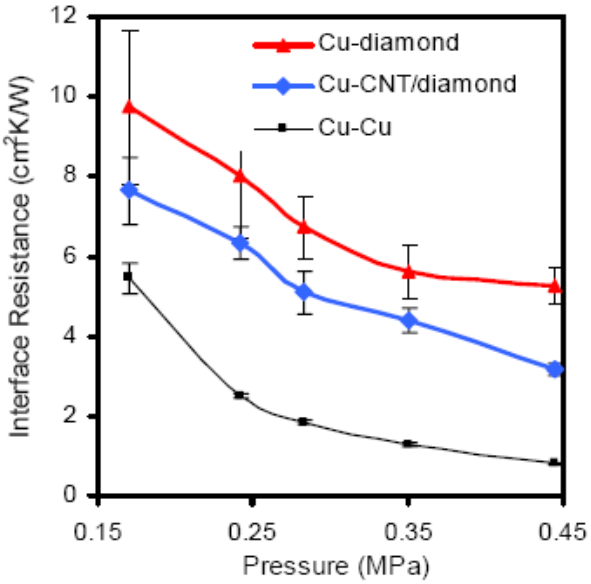

Figure 3. Thermal resistance as a function of pressure for copper-copper, copper-diamond interfaces and in combination with a CNT array (Cu-CNT/diamond).

SEM images of the copper bars' end surfaces after the test of Cu-diamond interface showed a very low density of contacted regions. The total contacted region at Cu-diamond interface is estimated to be less than 0.8% of the total interface area. A similar contact phenomenon was observed on the copper surface after the test of the Cu-CNT/diamond interface. An image from the post-experiment examination of the CNT-diamond composite indicates that only part of the CNT array conformed well to the topological profile of the copper bar.

The low contact area at the Cu-diamond and Cu-CNT/diamond interfaces explains the large values of measured thermal contact resistances. However, such a low contact area and gaps larger than the CNT array's height can not be accounted for solely by the surface roughness of the diamond film. A topological investigation of diagonal full-span scans over both sides of the double-side diamond film specimen revealed a significant deformation of the substrate. The surface profiles show that very large substrate deformations (>100μm) occurred near the outer edges of the chip. Apparently, diamond film contraction on one side of the double-side coated substrate dominated the deformation. Such deformation and consequent stress of the substrate chip resulted in the low contact area at the Cu-diamond and Cu-CNT/diamond interfaces. These

results suggest two possible methods to prepare flat diamond-coated samples. The first is to prepare a sample with larger dimension, and then trim it to the final size with laser cutting techniques so that the bent area will be removed. The second method is to grow the films on circular disc substrate in which edge effects should be less prominent.

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