Contents lists available at ScienceDirect

Acta Materialia
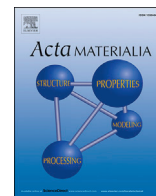
Full length article

Control of the in-plane thermal conductivity of ultra-thin nanocrystalline diamond films through the grain and grain boundary properties

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ARTICLE INFO

ABSTRACT

The in-plane thermal conductivity of polycrystalline diamond near its nucleation site, which is a key parameter to an efficient integration of diamond in modern high power AlGaN/GaN high electron mobility devices, has been studied. By controlling the lateral grain size evolution through the diamond growth conditions it has been possible to increase the in-plane thermal conductivity of the polycrystalline diamond film for a given thickness. Besides, the in-plane thermal conductivity has been found strongly inhomogeneous across the diamond films, being also possible to control this inhomogeneity by the growth conditions. The experimental results has been explained through a combined effect of the phonon mean free path confinement due the grain size and the quality of the grain/grain interfaces, showing that both effects evolve with the grain expansion and are dependant on the diamond growth conditions. This analysis shows how the thermal transport in the near nucleation region of polycrystalline diamond can be controlled, which ultimately opens the door to create ultra-thin layers with a engineered thermal conductivity, ranging from a few W/m K to a few hundreds of W/m K.

1. Introduction

Recently, the integration of diamond with high power AlGaN/GaN high mobility electron devices (HEMTs) was demonstrated to be a very promising solution to optimize their heat management [1–10], which enables handling much higher operational electrical power densities [1]. To take full advantage of the high thermal conductivity of diamond, reaching up to 3000 W/m K for single crystalline high quality diamond, the diamond heat dissipation layer should be located as close as possible to the heat source, i.e. the device channel [4,8]. This is achieved by directly growing diamond films on the devices, which results in polycrystalline diamond films rather than single crystal diamond. However, while the thermal conductivity of polycrystalline diamond may reach values close to that of the single crystal diamond [11], the thermal transport near its near nucleation site may be much lower due the small grain size and the accumulation of defects in this region [12,13]. Two strat-

egies for combining diamond with the devices using the direct growth approach have emerged in the recent years, namely, either by substituting the SiC or Si substrate [1,3–5], or by growing the diamond films on top of the device passivation layer [6–10]. However, in both strategies the heat has to diffuse across the nucleation region of the diamond film, and therefore knowing how the heat is spread in the first microns of the polycrystalline diamond is fundamental in order to optimize their thermal resistance and thus improve their lifetime and reduce its energy consumption. A few inconsistent results are available in the literature about the in-plane thermal transport in the first microns of polycrystalline diamond showing values ranging from a few W/m K up to 800 W/m K for layers thicknesses below 2 µm [13–24]. Also a strong inhomogeneity of the in-plane thermal conductivity through the diamond films has been reported [11,12,21,22,24], although mostly for films larger than a few micrometer thickness, due to challenges in measuring the thermal properties of very thin diamond films. Therefore a clear description of the heat transport in the complex near nucleation region of polycrystalline diamond is still lacking. In this paper, we study the in-plane thermal transport in the near nucleation region of polycrystalline diamond, which consist on nanocrystalline diamond

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(NCD), through a set of controlled ultrathin diamond films of different thickness, grain sizes and growth methods, with its thermal conductivity being determined with a direct steady-state technique developed recently [24]. This enables the correlation between the lateral heat transport and the physical properties of the samples, namely thickness and grain sizes. The results can be explained through a theoretical model which takes into account the effect of the grain size in terms of shortening of the phonon mean free path and also the temperature gradients in the defective grain boundaries. This analysis allows the characterization of the quality of the grain/grain interfaces and also explains the inhomogeneous in-plane thermal conductivity observed in ultra-thin diamond films.

2. Experimental details

NCD films were grown on single crystal Si <100> wafers using two different strategies, hot filament chemical vapor deposition (HF-NCD) and microwave plasma chemical vapor deposition (MW-NCD). The deposition of the NCD film on a foreign substrate is a 2 steps process consisting on a nucleation phase, followed by an overgrowth stage. The HF-NCD samples were nucleated under similar conditions to the ones described in Ref. [25] by bias enhanced nucleation (BEN) performed in-situ in the same reactor used for the overgrowth, resulting in crystallite densities of more than 10^{10} cm^2. For the MW-NCD samples the seeding process described in Ref. [26] was used to form the nucleation layer. For this, Si wafers were first sonicated in a nanodiamond-ethanol solution for 30 min; and then in pure ethanol to remove all non-adherent diamond seeds. The seeding solution is a suspension of detonation nanodiamond, with an average grain size of 4–6 nm. Next, wafers are placed in a spin tool where they are spray-rinsed and dried. This seeding method typically achieves uniform nucleation with a density exceeding 10^{12} cm^{-2}. After the nucleation, the grain evolution of diamond may be controlled during the overgrowth phase by varying the concentration of methane in the hydrogen carrier gas (CH$_4$/H$_2$), the temperature of the substrate (T$_{subs}$) and the pressure of the CVD chamber [27–29]. Two sets of growth conditions at 1.5 kPa were used to grow the HF-NCD films of different thicknesses: 470, 680 and 1000 nm using 0.4% CH$_4$/H$_2$, and a T$_{subs}$ of 750 °C (recipe A) and 480, 860, 980 and 1500 nm with 0.2% CH$_4$/H$_2$, and T$_{subs}$ = 825 °C (recipe B). The two growth conditions were chosen to produce a different lateral grain evolution, resulting in different grain sizes for a given thickness, illustrated in the scanning electron microscopy (SEM) micrographs in Fig. 1-a). Note that samples grown with Recipe B exhibit faster lateral grain expansion (see Fig. 1). The average in-plane grain size on the top of the diamond films was determined from the SEM micrographs using the three-circle procedure proposed by Abrams for polycrystalline materials [30], with the results shown in Fig. 1-b). Note that for the two HF-NCD recipes the lateral grain size follows a linear relationship with thickness, with the slope for the recipe B faster than the one for recipe A. This lateral expansion holds within the first 1000 nm, while thereafter the grain size lateral expansion is lowered (see Fig. 1-b sketch). To test the general applicability of the results and model obtained here through HF-diamond in a broader field, the results were compared to similar MW-diamond grown at a substrate temperature of 750 °C with a CH$_4$/H$_2$ ratio of 0.5 and 0.33 for recipes 1 and 2, respectively. Such conditions yield a similar lateral grain size at 1 µm as the HF-NCD recipes A and B respectively (Fig. 1-b).

For measuring the in-plane thermal conductivity of these ultra-thin NCD films, we applied a technique described in Ref. [24] which is based on Raman thermography assisted by Silicon nanowires acting as surface nano thermometers. For measuring Raman spectra a Renishaw InVia spectrometer with an Ar+ 488 nm laser line was used. A lateral resolution better than 1 µm was obtained by using the confocal mode with a 50× 0.65 NA objective. Free standing membranes were created in the samples by selectively etching away the silicon substrate underneath the diamond. Etching of the Si substrate was accomplished both by a wet-etch (MW-NCD) and by a dry-etch process (HF-NCD). In the former, the etching was performed by dipping the sample into a KOH solution while for the dry-etch process, etching was performed in a inductively coupled plasma (ICP) reactor by means of SF$_6$/C$_4$F$_8$ gases; for this a etch-mask of 300 nm thick Al film was used. On top of the free standing membranes, 5 µm wide thin metal stripe (Ti/Au) was deposited in the center of the free standing membrane to act as a heating source. When a current flows through the metal stripe, the Joule heating results in heat which flows into the diamond and laterally along the membrane, and ultimately into the Si substrate. Silicon NWs were also deposited on top of the membranes and metal surface providing an accurate local peak temperature measurement. From the temperature profiles measured from the heater to the edge of the substrate, lateral thermal conductivity can be extracted by solving the Heat equation. Temperature was measured in the diamond membrane and from the silicon NWs by means of Raman thermography, based on the phonon frequency shift as function of temperature as described in Ref. [24].

3. Experimental results

Fig. 2 shows an example of a temperature profile obtained from the diamond membranes for the 470 nm thick HF-NCD of recipe B, and corresponding temperature profiles are obtained for the other investigated samples. The determined in-plane thermal conductivity of each as-grown NCD layer is shown in Fig. 3. The thermal conductivity values are in general one to two orders of magnitude below the thermal conductivities reported for bulk CVD diamond for all the investigated samples [11]. Both families of HF-NCD samples show a linear correlation with its layer thickness (see Fig. 3-a). We also noted that the slope of the temperature profiles used to determine the thermal conductivity can be fitted assuming a temperature independent in-plane thermal conductivity. In bulk diamond, and above room temperature, it is well known that the thermal conductivity decreases with temperature due to the 3 phonon scattering mechanism [31], being this decay well reproduced by a power law given as $k(T) = k_{300\,K}(T/300\,K)^{-1.25}$ extracted from the fitting to the experimental data of Ref. [32]. However, when such temperature dependence is assumed for the NCD membranes it is not possible to simultaneously reproduce the experimental temperature profiles for different powers dissipated in the heater (see Fig. 2). This deviation from the bulk behavior of the phonon transport indicates that the in-grain phonon phonon scattering in the near nucleation site of the NCD membranes is not the dominant phonon scattering mechanism in these samples from room temperature and above, being the dominant phonon scattering mechanism either due to the phonon scattering at the grain boundaries or the phonon scattering due defects/impurities in the lattice.

The thermal conductivity of samples of recipe B are higher than those samples grown with recipe A, due to the different evolution of the grain size (Fig. 1-b). The results obtained for the MW-NCD samples are consistent with the HF-NCD. The importance of the grain size for thermal conductivity is highlighted in Fig. 3-b, where the in-plane thermal conductivity as function of the average grain size measured at the surface of the diamond films is displayed. Independent of growth recipe, thickness and growth method a nearly linear correlation between the in-plane thermal conductivity and the average grain size is observed. These results illustrate that as a thumb rule the as-grown NCD samples with a faster lateral grain