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Copper induced synthesis of graphene using amorphous carbon

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ABSTRACT

Copper is approaching its reliability limits with respect to electromigration due to very high current density as a result of continuous technology scaling. Graphene on the other hand has excellent electrical and thermal properties which can prove to be a vital candidate for improving the reliability performance of copper interconnections in ULSI. Possibility of crystallization of amorphous carbon into graphene catalyzed by copper thin film is demonstrated in this work, as evidenced by the Raman, XPS and SIMS analysis, and the number of graphene layer synthesized can be modified with the method developed. As the synthesized graphene layers are on top of the copper film whilst the amorphous carbon source is below the copper film, no contamination of the graphene layer is presence with the method developed, improving the quality and uniformity of the grown graphene layers.

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1. Introduction

Graphene is a material of great interest today due to its magnificent properties such as single atom thickness [1], high current density tolerance [2], extremely high mobility [3] and high thermal conductivity [4], to name a few.

Various methods to obtain graphene has been proposed since its discovery in 2004 [1], starting from mechanical exfoliation of graphite using a tape [1] to the currently used chemical vapor deposition (CVD) technique [5–8]. A simpler low cost technique to obtain non-transferable graphene using amorphous carbon as the solid source at elevated temperature, with nickel or cobalt thin film as catalyst has been reported [9,10]. However, such graphene synthesis method was claimed to be impossible with copper due to the very low solubility of carbon in copper [9,10].

On the other hand, since the applications of copper material is vast and important, such as ULSI interconnects, printed circuit boards, transmission wires and cables, mechanical chassis, automobile spare parts, aviation equipment etc., and that several reliability concerns associated with copper have been reported [11–13] which poses limitations (such as ease of corrosion and oxidation, limited electromigration lifetime etc.) on its usage, the growth of graphene on copper can overcome some limitations of copper materials. Presence of graphene on copper can also push the electrical and thermal conductivity limits of copper to higher values and provide better electromigration [12,13] performance.

Growth of graphene on copper using chemical vapor deposition (CVD) has been demonstrated by Li et al. [14,15] and others [16–18]. However, these methods use expensive carbon source which is also wasted during chamber cleaning, and they also have no control over number of layers of synthesized graphene. In view of the limitation of the reported methods, the use of amorphous carbon as solid source for graphene synthesis on copper should be explored because it is economical and can produce pure carbon species responsible for graphene growth. Ji et al. [19] successfully demonstrated this possibility but their method involved graphene deposition on a copper foil which is several microns thick, and the graphene layers so obtained needs to be transferred on the required substrate which might introduce defects. Also the amorphous carbon layer atop copper crystallizes to graphene in their method, thus the graphene is in direct contact with amorphous carbon, renders the possibility of crystal contaminations in the synthesized graphene layers. Such crystal contaminations are undesirable as it is considered to be one of the reasons for low electron mobility of CVD graphene which is a consequence of growth mechanism of graphene on copper [20–21]. In fact, Ruiz et al. [22] demonstrated the negative effects of contamination on the graphene's quality and uniformity.

In this work, we demonstrate the feasibility of crystallization of amorphous carbon below sputtered copper thin film which acts as catalyst to obtain graphene at the top surface of the copper film experimentally. The growth mechanism of the method developed here will be reported elsewhere.

2. Experiment

2.1. Preparation of samples

The experiment involves deposition of amorphous carbon (a-C) thin film and copper (Cu) (99.99%) film of thickness 60 nm and 800 nm

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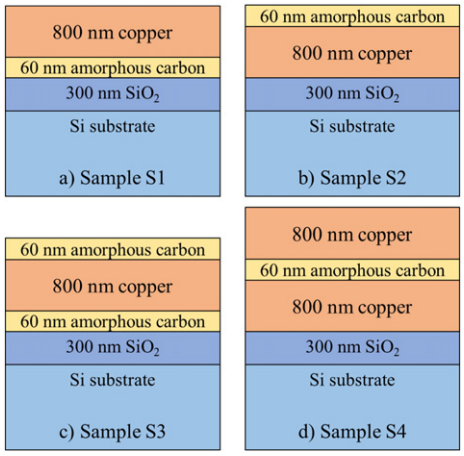

Fig. 1. Structure configurations of the samples for experimentation in this work; a) sample S1 with 60 nm a-C layer underneath 800 nm Cu thin film, b) sample S2 with 60 nm a-C layer atop 800 nm Cu thin film, c) sample S3 with 800 nm Cu thin film sandwiched between two 60 nm a-C layers and d) sample S4 with 60 nm a-C layer sandwiched between two 800 nm Cu thin films.

respectively on Si/SiO_2 (300 nm) substrate with different configurations as shown in Fig. 1. The deposition is carried out using RF and DC sputtering for a-C and Cu respectively at a stable pressure of 3 mTorr in the presence of Ar gas with a flow rate of 30 sccm. The substrate temperature during the sputtering is maintained at 250 °C.

2.2. Annealing

Subsequently, all the samples are annealed in hydrogen (H_2) environment with a flow rate of 50 sccm at a low pressure of 1 Torr and annealing temperature of 1020 °C for 50 min. After annealing, the samples are cooled down during which the H_2 flow rate is decreased to 30 sccm and argon (Ar) gas is introduced at a flow rate of 20 sccm.

3. Results & discussion

3.1. Raman characterization

Fig. 2 shows the Raman spectra (measured with a Raman laser of 473 nm) of sample S1 after annealing. A clear evidence of the presence of multi-layer graphene can be seen from the 2D peak with I_{2D}/I_G and

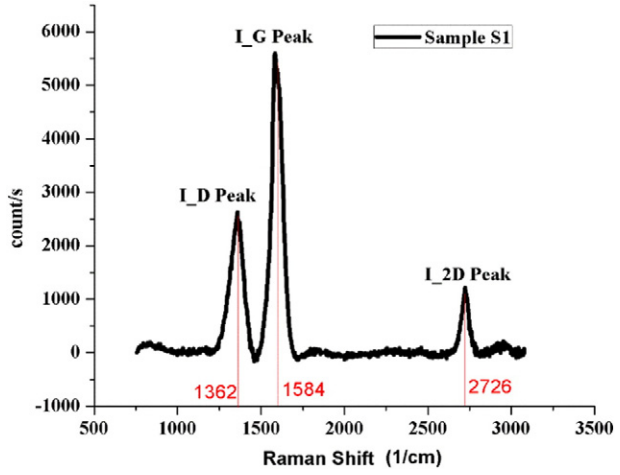

Fig. 2. Raman spectrum of annealed sample S1; I_D peak appears at wave number 1362 cm^{-1}, I_G peak appears at wave number 1584 cm^{-1} and I_{2D} peak appears at wave number 2726 cm^{-1}. This Raman signature belongs to multi-layer graphene.

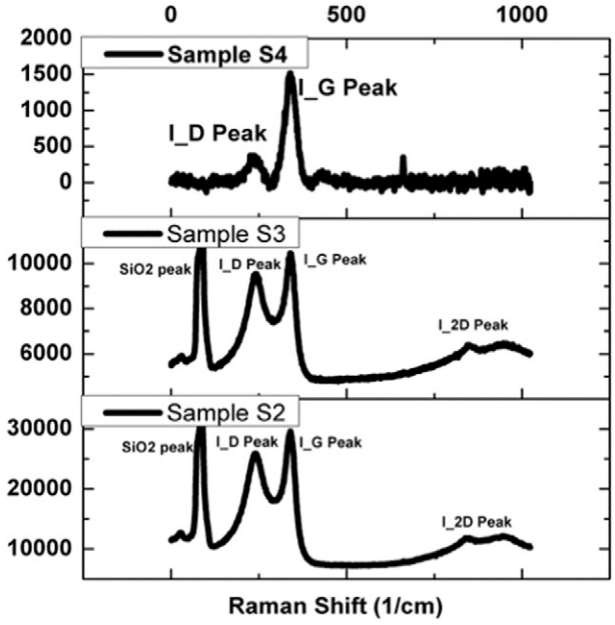

Fig. 3. Raman spectrum of annealed samples S2, S3 and S4. The Raman signature for samples S2 and S3 appears to be somewhat different from graphene's signature while that of sample S4 is more close to disordered graphite. None of these configurations have evidence of the presence of graphene.

I_D/I_G ratios of 0.29 and 0.49 respectively, showing that the crystallization of a-C into graphene catalyzed by Cu is feasible.

Fig. 3 shows the Raman spectra of samples S2, S3 and S4, and only visible but very low I_{2D} peak and significant but peculiar I_G and I_D peaks are found for samples S2 and S3. The underlying reasons are currently under investigation.

On the other hand, no I_{2D} peak is visible for sample S4. In sample S4, both the copper layers are available as catalysis site for initiation of carbon crystallization, and graphitization of a-C is more likely than the formation of hydrogenated tetrahedral amorphous carbon (T a-C:H) [23] on the top surface. Consequently, the crystallization is rapid and by the end of 50 min of annealing time, the H_2 gas etches off almost all graphene, leaving behind disordered graphite [24].

It is suspected that the low intensity 2D peak in sample S1 could be due to the presence of many graphene layers. To obtain fewer layers, samples S1.1 and S1.2 are prepared which are similar in configuration

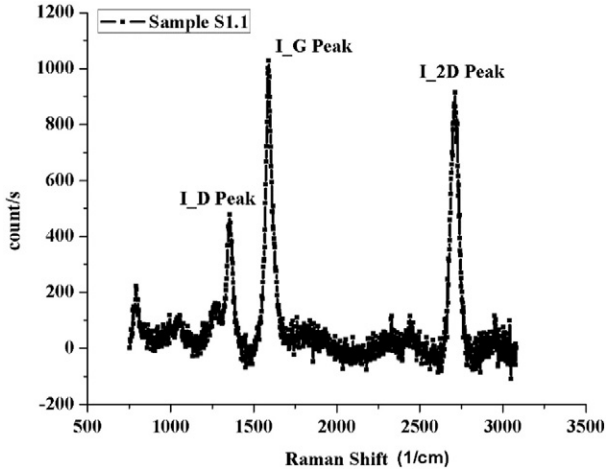

Fig. 4. Raman signatures for samples S1.1; I_D peak appears at wave number 1355 cm^{-1}, I_G peak appears at wave number 1589 cm^{-1} and I_{2D} peak appears at wave number 2711 cm^{-1}. This Raman signature corresponds to few-layer graphene.

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