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Evaluation of Acoustic Properties of Multilayer Graphene Sheet by Ultrasonic Microscopy

超音波顕微鏡による多層グラフェン薄膜の音響特性評価

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1. Introduction

Graphene is attracting a lot of interests due to its outstanding electronical properties such as high carrier mobility. In addition, its mechanical and electrochemical features have motivated many researchers to study nano/micro electro mechaical systems (N/MEMS) and sensor applications of graphene.[1)] For such works, "Scotch tape technique" is widely used to obtain a graphene monolayer. Chemical vapor deposition (CVD) is another technique, but the available graphene quality is still limited.

Murakami *et al.* synthesized a high quality and highly oriented multilayer graphene sheet (MGS) by annealing a polymide film.[2)] This sheet is attractive for electromechanical applications, e.g. sensors, rather than semiconductor ones. However, the electromechanical properties have not been sufficiently revealed. In this study, therefore, we investigated elastic stiffness constants of a MGS from acoustic measuement and numerical calculation.

First, the phase velocity of leaky surface acoustic wave (SAW) on the MGS was measured using a line-focus-beam ultrasonic material characterization (LFB-UMC) system.[3,4)] Then, theoretical calculation was conducted to determine the material constants based on the measurement results.[5)] In addition, the velocity and electromechanical coupling coefficient, k^2, of 0-th symmetric (S_0) mode Lamb wave on ZnO/MGS were calculated by finite element method (FEM) using the determined constants.

2. Experimental method

A MGS was prepared as follows. First, a polyimide film of 27 μm thickness was pyrolyzed and carbonized for 20 minutes at 950ºC in N_2 atmosphere. Next, the specimen was annealed for 30 minutes at 2800ºC in Ar atmosphere for graphitization. As a result, a MGS of 11 μm thickness was obtained. Figure 1 is the

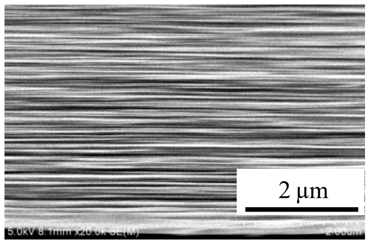

Fig. 1 Cross-sectional SEM image of MGS.

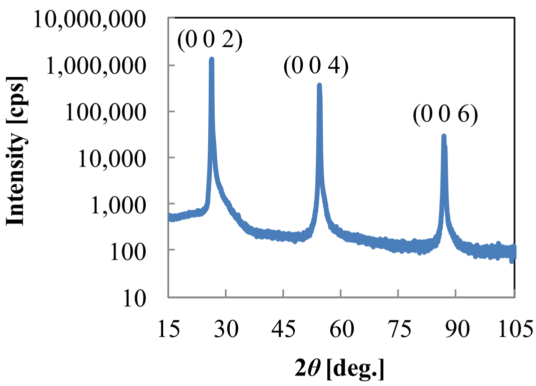

Fig. 2 XRD θ-2θ spectrum of MGS.

cross-sectional scanning electron micrograph (SEM) image, showing that the specimen is composed of multilayer graphene without voids. Figure 2 shows the X-ray diffraction (XRD) spectrum of the MGS. The sharp (002) peaks are derived from the layered structure, and the full width of half maximum (FWHM) of (002) is as small as 0.947°.

The MGS was diced into 10 mm square samples, and the sample was attached to a c-plane sapphire substrate as shown in Fig. 3. First, the sapphire substrate was cleaned by excimer irradiation to remove organic contaminations, and then spin-coated with UV bond (TB3042B, ThreeBond) (Fig. 3 (a)). Next, the MGS and a dummy Si substrate were pressed at 0.25 kN and exposed to UV for 300 seconds using a UV curing machine (UMA-802, USHIO) (Fig. 3 (b)). After that, the dummy Si substrate was removed manually (Fig. 3 (c)). The thickness of the UV bond is approximately 0.1 μm.

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Leaky SAW was generated on the MGS/UV bond/sapphire sample using a ZnO transducer of the LFB-UMC system, and simultaneously the phase velocity was measured. The detail is found elsewhere.[3,4]

3. Results

Figure 4 shows the velocity of leaky SAW versus MGS thickness normalized by wavelength, λ. The velocity was 5409-5661 m/s at 170-270 MHz.

To determine material constants, we extended Campbell's method to leaky SAW on the MGS/UV bond/sapphire structure.[6] All the constants were determined to minimize error between the measured and calculated velocities. As a result, the constants of the MGS were calculated as c_{11}^E = 1060 GPa, c_{13}^E = 14.9 GPa, c_{33}^E = 36.4 GPa, c_{44}^E = 3.50 GPa, and c_{66}^E = 440 GPa.

Using the obtained constants, the velocity and k^2 of S_0 mode Lamb wave on ZnO/MGS were calculated by FEM. Note that the constants of ZnO and c_{12}^E of the MGS were from references.[7,8] Figures 5 and 6 show the calculated velocity and k^2 of S_0 mode on ZnO/MGS at different MGS thickness, respectively. Both velocity and k^2 increase by the MGS compared to those on a ZnO monolayer, if the thickness of ZnO, h, is thinner than 0.3λ. This result suggests a potential of the MGS as a medium of high frequency SAW devices.

4. Conclusion

The material constants of a MGS were determined by acoustic measurement and numerical calculation. The velocity of leaky SAW on MGS/sapphire was measured as 5409-5661 m/s at 170-270 MHz by the LFB-UMC system. We also suggested a possibility of a ZnO/MGS structure as high frequency SAW devices by FEM.

Acknowledgment

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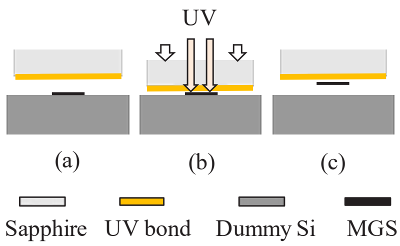

Fig. 3 Bonding process of MGS and sapphire substrate. (a) specimen set up. (b) Pressing and exposing UV. (c) dummy Si substrate removal.

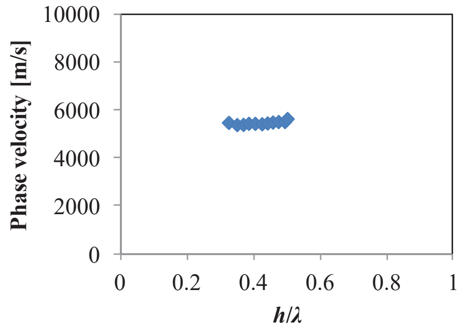

Fig. 4 Phase velocity of leaky SAW versus MGS thickness.

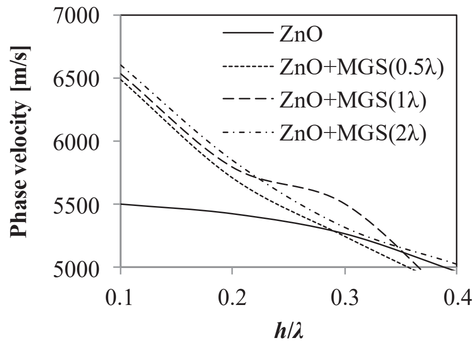

Fig. 5 Phase velocity of S_0 mode on Lamb wave versus ZnO thickness.

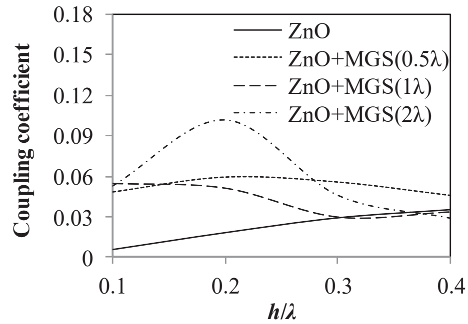

Fig. 6 Electromechanical coupling coefficient of S_0 mode Lamb wave versus ZnO thickness.