Film Structures of Fe/B-doped Carbon/Fe₃Si Spin Valve Junctions

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(Received January 24, 2017)

Fe/B-doped carbon/Fe₃Si trilayered films were prepared on Si(111) substrates by physical vapor deposition with a mask method, and the film-structures and magnetic properties of the films were investigated. The Fe₃Si and Fe layers were deposited by facing targets direct-current sputtering (FTDCS), and the B-doped carbon layers were deposited by coaxial arc plasma deposition (CAPD) with B-blended graphite targets. Here, since the B-doped carbon layers were deposited by CAPD in the same manner as the deposition of B-doped ultrananocrystalline diamond/hydrogenated amorphous carbon composite (UNCD/a-C:H) films, the interlayers should be B-doped UNCD/a-C:H. The formation of a layered structure was confirmed by transmission electron microscopy (TEM). The diffusion of Fe and Si atoms into the interlayer occurs in the range of several nanometers. The shape of the magnetization curve has clear steps that evidently indicate the formation of antiparallel alignment of magnetizations owing to the difference in the coercive forces between the top Fe and bottom Fe₃Si layers. It was experimentally demonstrated that B-doped UNCD/a-C:H is applicable to Fe-Si system spin valves as interlayer materials.

1. Introduction

Spintronics devices that utilize both electric charge and spin of electrons have actively been studied. Giant magnetoresistance (GMR) [1,2] and tunnel magnetoresistance (TMR) [3-8] effects are representative phenomena utilizing the spin-dependent scattering of carriers.

Whereas GMR and TMR films employ nonmagnetic metal and insulator as interlayers, respectively, there have been a few studies on spintronics devices comprising semiconductors. Spin injection from ferromagnetic materials into semiconductors such as Si, Ge and GaAs, has been widely investigated thus far [9-14]. Recently, spin injection into semiconducting graphene has received much attention because of a long spin diffusion length being expected due to weak spin-orbit interactions [15-17].

Our laboratory has structurally and electrically investigated ultrananocrystalline diamond (UNCD)/hydrogenated amorphous carbon (a-C:H) composite (UNCD/a-C:H) films, comprising a large number of nano-sized diamond grains and an a-C:H matrix, prepared by coaxial arc plasma deposition (CAPD), thus far [18-22]. The UNCD/a-C:H films prepared by CAPD have the following characteristics: (i) the production of p and n-type conduction accompanied by enhanced electrical conductivities is possible by B and N doping, respectively; (ii) they can be grown on foreign solid substrates; (iii) they can be grown at low substrate temperatures even on unheated substrates, while
chemical vapor deposition (CVD) requires high substrate temperatures of more than 700 °C for the growth [23,24]. UNCD/a-C:H is a carbon-based semiconductor similarly to graphene, and it is a new candidate as spin transport materials.

We have studied Fe-Si based artificial lattices and spin valves comprising ferromagnetic Fe$_3$Si and semiconducting FeSi$_2$, prepared by sputtering, thus far [25-36]. Based on the preparation techniques in our previous researches, spin valve junctions comprising ferromagnetic Fe$_3$Si and Fe layers and B-doped UNCD/a-C:H interlayers were prepared, and they were structurally investigated by transmission electron microscopy (TEM). We report that it is possible for layered structures to be formed owing to the low substrate temperature growth of B-doped UNCD/a-C:H interlayers.

2. Experimental methods

Fe (100 nm)/B-doped ultrananocrystalline diamond (10 nm)/Fe$_3$Si (100 nm) trilayered spin valve junction were deposited on p-type Si(111) substrates by a mask method, as shown in Fig. 1(a). First, the Si(111) substrate with specific resistance range of 3000–8000 Ω·cm, produced by a floating zone (FZ) method, was cleaned with 1 % hydrofluoric acid and rinsed in deionized water before it was set into the vacuum chamber of a facing target direct-current sputtering (FTDCS) apparatus. The bottom Fe$_3$Si layer (100 nm) was deposited on the Si(111) substrate at an Ar pressure of 1.33 × 10$^{-1}$ Pa and substrate temperature of 300 °C, by FTDCS using the 1st mask. Subsequently, after the exposure to air for the mask exchange, the UNCD/a-C:H interlayer (10 nm) was deposited with the 2nd mask at a H$_2$ pressure of 53.3 Pa and substrate temperature of 300 °C by CAPD with a 10 at.% B-blended graphite target. After the exposure to air for the mask exchange again, the top Fe layer (100 nm) was deposited with the 3rd mask at an Ar pressure of 1.33 × 10$^{-1}$ Pa and room temperature by FTDCS apparatus. The optical top-view image of the resultant junction is shown in Fig. 1(b). The B content of the UNCD/a-C:H interlayer was estimated to approximately 5 at. % from the X-ray photoemission spectra. The microstructure of the trilayered films were structurally observed by TEM combined with energy dispersive X-ray (EDX) spectroscopy. The magnetization curves of the junctions were measured at room temperature using a vibrating sample magnetometer (VSM).

3. Results and discussion

Figure 2 shows the XRD pattern of the junction, measured in 2θ-θ. The junction exhibit diffraction
peaks of Fe$_3$Si-222 and Fe-110 and 200. In our previous study, we confirmed that Fe$_3$Si oriented grains are also in-plane ordered on Si(111) substrates, by X-ray diffraction (XRD) [35,36]. From a pole figure pattern concerning the Fe$_3$Si-422 plane with a rotation axis of Fe$_3$Si [222], the in-plane orientation of the Fe$_3$Si layer was confirmed. Totally considering this results and our previous research results [26], the bottom Fe$_3$Si layer is epitaxially grown on the Si(111) substrate. Since the top Fe layer exhibits Fe-110 and 220 peaks and it is deposited on the non-oriented UNCD/a-C:H layer, the Fe layer should be non-oriented polycrystalline. Here, the Fe-110 peak might be buried in the background pattern of the Si substrate.

Figure 3(a) shows the cross-sectional high-angle annular dark field scanning transmission electron microscopic (HAADF-STEM) image of the junction. A trilayered structure is certainly formed, in other words, the UNCD/a-C:H layer structurally acts as an interlayer that clearly separates the top Fe layer from the bottom Fe$_3$Si layer. Figure 3(b) shows a bright-field scanning transmission electron microscopic (BF-STEM) image of an Fe$_3$Si/Si(111) interface in the junction. The interface is not sharp in atomic scale, which might be because of atomic interdiffusion induced by the bombardment of sputtering species and the HF pretreatment of the Si substrate was imperfectly made.
From the electron diffraction pattern, the epitaxial growth of the Fe$_3$Si layer on the Si(111) substrate was confirmed, as expected from the XRD measurement results. This is consistent with results in our previous research, wherein Fe$_3$Si thin films are epitaxially grown on Si(111) substrate even at room temperature [26].

The cross-sectional HAADF-STEM image of a magnified area around the interlayer of the junction and the EDX profile of C, O, Si, and Fe in the depth direction are shown in Fig. 4(a) and 4(b), respectively. The STEM image of the interlayer can be divided into three areas. At the side areas of A and B, O atoms preferentially exist, which means that the interfaces are oxidized due to temporal exposures to air for the replacements of the masks. Additionally, C, Si, and Fe atoms coexist in the areas of A and B. This is because of the diffusions of Fe and Si atoms into the interlayer. Surprisingly, the center area between the A and B areas hardly contains all atoms. The TEM observation sample was prepared by a focused ion beam (FIB) apparatus. As a probable reason for it, we consider that the center C-rich areas were preferentially etched away by Ga ions bombardment during the FIB process. Since the interlayer exists in layer structure, the UNCD/a-C:H layer should have been present before the FIB etching.

The magnetization curve of the junction measured at room temperature is shown in Fig. 5. The magnetization curve has clear two steps that indicate the antiparallel alignment formation of
magnetizations owing to the difference in the coercive forces between the top Fe and the bottom Fe$_3$Si layers. The polycrystalline Fe layer has a larger coercive force than that of the epitaxial Fe$_3$Si layer [37]. The spin valve behavior is realized, which proves that the UNCD/a-C:H layer act as an interlayer for the spin valve action.

4. Conclusion

Trilayered junctions comprising Fe, B-doped UNCD/a-C:H, and Fe$_3$Si layers were prepared on Si(111) substrates by physical vapor deposition combined with a mask method, and the film-structures and magnetic properties were investigated. The TEM observation revealed that the interlayer exists in layered structure and it contains oxidized layers at interfaces with ferromagnetic layers due to the exposure to air for the replacements of the masks. From the shape of the magnetization curve, the formation of antiparallel alignment of magnetizations owing to the difference in the coercive forces between the top Fe and bottom Fe$_3$Si layers were confirmed. It was experimentally demonstrated that UNCD/a-C:H is applicable to Fe-Si system spin valves as interlayer materials although the diffusion of Si and Fe atoms from the ferromagnetic layers into the interlayer occurs in the range of several nanometers.

Acknowledgment

This work was partially supported by JSPS KAKENHI Grant Number JP16K14391, JP15K21594, Yoshida Grant from Yoshida Academic Education Promotion Association, and Tohoku University Advanced Characterization Nanotechnology Platform in Nanotechnology Platform Project sponsored by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

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