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Planar Hall Voltage Properties of Ta/NiFe/Ta/NiFe/Ta Multilayer for Biosensor Applications

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Recently, magnetic nanoparticles and planar Hall resistance (PHR) devices used in biomolecule diagnosis and biological applications have attracted considerable attention as biosensors. The voltage properties of a planar Hall device with a Ta/NiFe/Ta/NiFe/Ta multilayer that was prepared in a cross-shaped pattern at a width of 0.5 mm using a shadow mask, were investigated. The sensitive planar Hall voltage (SPHV) obtained from the planar Hall voltage (PHV) curves of a Ta(5 nm)/NiFe(6 nm)/Ta(t = 2-8 nm)/NiFe(6 nm)/Ta(5 nm) multilayered sample was a few mV/Oe in the fine magnetic field region of the external magnetic field of ± 1.0 Oe. In particular, the thickness of Ta(t = 8 nm), the PHR ratio, magnetic sensitivity, and SPHV improved significantly by as much as 36.4 %, 28 %/Oe, and 1.6 mV/Oe, respectively. These results can be used in the development of a PHR biosensor with a high sensitivity for the detection of nanoparticle-antibody binding, even in a simple structure formed only out of a ferromagnetic NiFe layer.

Keywords : external magnetic field, planar Hall resistance (PHR), sensitive planar Hall voltage (SPHV), magnetic nanoparticle, antibody

1. Introduction

Magnetic nanoparticles are classified into pure metals, metal oxides, and magnetic nanocomposites [1]. Magnetic iron oxide nanoparticles are largely composed of two types of iron oxide cores (magnetite Fe_3O_4 and maghemite γ-Fe_2O_3), and have a core-shell structure containing a surface modified with various types of biomolecules, such as carbohydrates, polymers, lipids, and proteins [2]. Traditionally, magnetic nanoparticles have been effectively used to separate or purify DNA, proteins, and peptides along with magnetic microparticles. Magnetic nanoparticles exhibit superparamagnetism, and have magnetic properties only in the presence of an external magnetic field. When various ligands are introduced after stabilizing the surface of magnetic nanoparticles with lipids or polymers, DNA/RNA, proteins, bacteria, viruses, and cancer cells can be effectively separated using only a simple magnet [1].

The use of magnetic nanoparticles and beads in biomedical applications has been a major topic of interest. The magnetic nanoparticles widely used in the biomedical field are Co, Fe, Ni, Ti, iron oxide, and some ferrites. Fe_2O_3 or Fe_3O_4 nanoparticles are commonly used because of their low toxicity [1, 2]. As biomolecules link magnetic nanoparticles to biochemical reactions, the behavior of the particles and their ability to treat magnetic hyperthermia can be greatly improved when they are used as external magnetic pulse stimulation [3, 4]. Reports on the detection of magnetic nanoparticles bound to biomolecules composed of erythrocytes or antibodies of target cells are emerging [5, 6]. Currently, there is a need for a biosensor that can accurately measure the strength of the fine magnetic field emitted from multiple magnetic beads or magnetic nanoparticles [1, 2, 6].

In magnetic multilayer thin films showing the Hall effect, studies on increasing the sensitivity of the planar Hall resistance (PHR) for biosensor applications with magnetic nanoparticles have been conducted [7-9]. The PHR is caused by the magnetic anisotropy effect due to the change in the external magnetic field applied horizontally. This PHR sensor is mainly based on antiferromagnetic material and has a two-layer thin film or a

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multilayer structure with a giant magneto-resistance spin valve (GMR-SV) [10-12]. To improve the magnetic sensitivity (MS), by considering the characteristics of the ferromagnetic layer in the antiferromagnetic IrMn-based GMR-SV multilayer structure, the thickness of the Cu layer between the antiferromagnetic and ferromagnetic layer or between the two ferromagnetic layers should be optimized [11]. The PHR sensor has the disadvantages of the usage of four terminals and low output voltage. Especially, in bio-label detection in liquid environment, the most important factor is the thermal stability of sensor, which is 1,000 times less compared with another MR sensors [13]. However, PHR biosensor has the advantages of possessing a high signal-to-noise ratio and low offset voltage, as well as a simple fine pattern [8].

This study compared the sensitivity characteristics of the PHR device for the Ta/NiFe/Ta/NiFe/Ta multilayered structure. As a device that measures fine magnetic field to be used as a biosensor, a simple multilayered cross-shaped current terminal and voltage terminal were formed. The planar Hall voltage (PHV) curve was measured at a 90° angle, with the easy magnetization axis in the horizontal direction on the thin film sample surface with the external magnetic field direction. The PHR ratio and sensitive planar Hall voltage (SPHV) were compared with the measured values.

2. Experiment Method

For thin film samples of magnetic and non-magnetic materials, a 450 μm thick p-type Si wafer, doped with boron to form a 300 nm oxide layer (SiO_2), was used as the substrate. The thin film was fabricated using a DC magnetron sputtering system [9]. The deposition system installed in the high vacuum chamber has two 3-inch targets and magnetron-sputtering gun devices separated from each other. It was installed so that the substrate could be placed between the permanent magnets on both sides during thin film deposition. In the sample on which the thin film was deposited, the magnitude of the magnetic field at the center was 350 Oe [10].

Figure 1(a) shows the thin film structure of Ta(5 nm)/NiFe(6 nm)/Ta(t = 0, 2, 4, 8 nm)/NiFe(6 nm)/Ta(5 nm), in which Ta and NiFe were sputtered with two targets of Ta and NiFe. A cross-shaped electrode with a line width of 0.5 mm was formed, as shown in Fig. 1(b), during thin film deposition using a brass plate metal shadow mask with a thickness of 0.1 mm [5]. Fig. 1(b) shows the actual shape of the thin film sample at the center, which is the intersection point of the cross-shaped pattern. As shown in Fig. 1(b), uniaxial anisotropy was induced by deposition under a magnetic field to facilitate magnetization in the x-axis direction of the patterned thin film sample. To minimize the change in magnetoresistive properties due to the oxide film and surface contamination of the thin film sample, indium was pressed using an ex situ method to form an electrode immediately [12].

The voltage terminal of the patterned thin film sample maintained a measurement method that was parallel to the current terminal. The sensing current applied to the current terminal was set at 10 mA. The direction of the external magnetic field applied on the horizontal plane of the thin film sample, when measuring the PHV of several tens to hundreds of μV and millivolts, is perpendicular to the voltage terminal supplied to the specimen, as shown in Fig. 1(b) [9, 12].

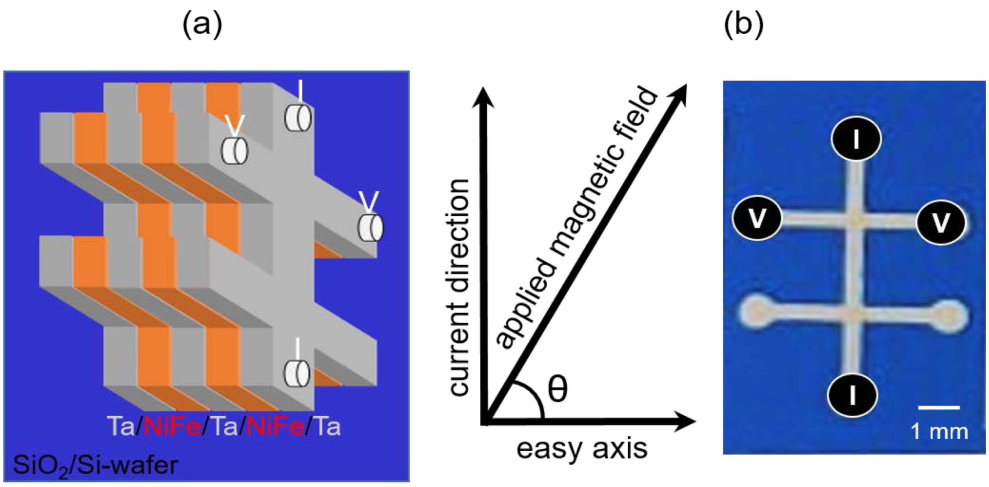

Fig. 1. (Color online) (a) The schematic structure of Si-wafer/SiO_2/Ta(5 nm)/NiFe(6 nm)/Ta(0-8 nm)/NiFe(6 nm)/Ta(5 nm) multilayer. (b) The angle between the magnetic easy axis and the applied magnetic field same to the current direction and photo image of two crosses patterned sample by using a shadow mask having a width of 0.5 mm.

The strength of the applied magnetic field, to obtain the PHV curve, was ± 10 Oe. In addition, the frequency of the function generator that determines the number and resolution of the measured data was 1.7 mHz. The PHV curve was obtained by fixing the sample at a 90° rotation angle (θ) along the horizontal plane of the thin film. In addition, the sensitivity was measured and analyzed by calculating the magnetic field sensitivity from the PHV curve measured between +10 Oe and −10 Oe, using a system that measures the current and voltage with a 4-terminal [12].

3. Results and Discussion

In Fig. 2, the magnetoresistance curve showed a major effect of anisotropic magnetoresistance (AMR) on spin–orbit coupling, in which the specific resistance of the thin film sample depends on the current direction, the easy axis of magnetization, and the direction of the external magnetic field [14]. In the Ta(5 nm)/NiFe(6 nm)/Ta(4 nm)/NiFe(6 nm)/Ta(5 nm) multilayer formed from the Ta layer, which is the bottom layer and the spacer layer inserted between the two ferromagnetic NiFe layers, the PHV curves are shown in Fig. 2(a) and Fig. 2(b). The thickness of the nonmagnetic Ta layer, between the two ferromagnetic layers, was 4 nm, the thickness of the ferromagnetic NiFe layer was 6 nm. The PHV curve, measured in the range of ± 10 Oe in the presence of an external magnetic field, showed a characteristic symmetrical curve based on 0 Oe. The coercive force of the upper and lower layers measured at room temperature was 2.2 Oe with the same thickness. The interlayer coupling did not occur and the coupling intensity between the lower and upper layer was 0 Oe.

The PHV curves, measuring the sensing current as 10 mA, showed a linear hysteresis pattern in the vicinity of 0 Oe, as shown in Fig. 2. The highest magnetic field sensitivity was in the region of ± 1.0 Oe and the PHR ratio calculated from the PHV curve at room temperature is 30.7 %. It showed a sensitivity characteristic with little hysteresis. Fig. 2(a) shows a PHV from +10 Oe to −10 Oe in an external magnetic field, showing linear sensitivity with 13.0 %/Oe near 0 Oe. Fig. 2(b) shows the PHV curve returning to the magnetic field region outside the range of coercive force from +10 Oe to −2 Oe, maintaining a PHR of 30.7 %. However, the hysteresis phenomenon showed asymmetrical characteristics at approximately 0 Oe. Alternatively, Fig. 2(b) shows the PHR characteristics due to the AMR effect of the NiFe layer separated by the thickness of Ta 4 nm, while maintaining the magnetic properties as the coercive force formed in the multilayered thin film sample was 2.2 Oe. Because the PHV curve in Fig. 2(b) at around 0 Oe changes along a reversible path, it displays a characteristic of linearly high sensitivity at 0 Oe without an external magnetic field, so that it can function as a biosensor. SPHV, having the same property as MS, is defined as the ratio of change in the PHV (ΔPHV) in relation to the magnetic field change in the linear region (DH), as shown in Fig. 2(b) [12]. For the Ta(5 nm)/NiFe(6 nm)/Ta(4 nm)/NiFe(6 nm)/Ta(5 nm) multilayer, the calculated value of SPHV is 800 mV/Oe.

The Ta(5 nm)/NiFe(6 nm)/Ta(t = 2 nm and 8 nm)/NiFe(6 nm)/Ta(5 nm) multilayer structure samples shown in Fig. 3(a) and Fig. 3(b) were set such that the current terminal connected around the right-angled cross-shape

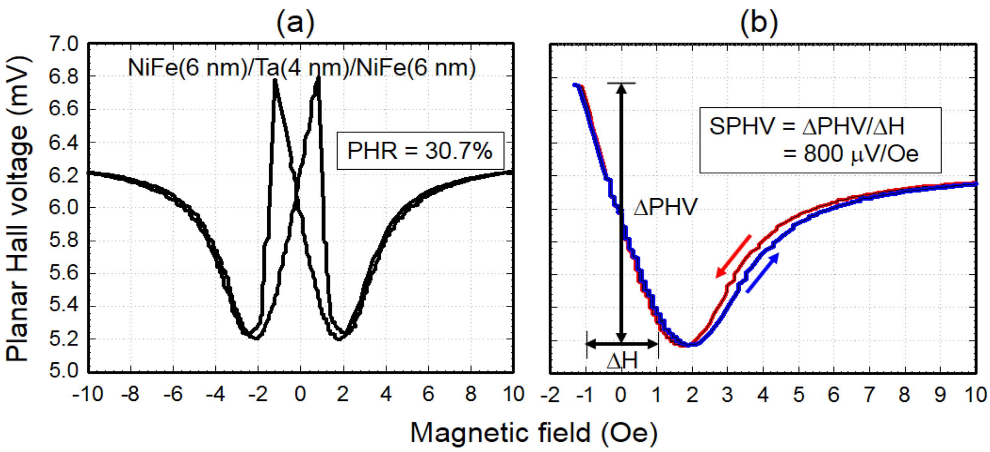

Fig. 2. (Color online) PHV curve for the Ta(5 nm)/NiFe(6 nm)/Ta(4 nm)/NiFe(6 nm)/Ta(5 nm) multilayer measured at the sensing current of 10 mA temperature in external magnetic fields of (a) −10 Oe ~ +10 Oe and (b) −2 Oe ~ +10 Oe. In the neighboring region of the external magnetic field of ± 1.0 Oe, the values of PHR ratio, MS, and SPHV (= ΔPHV/ΔH) are 30.7 %, 13.0 %/Oe, and 800 μV/Oe, respectively.

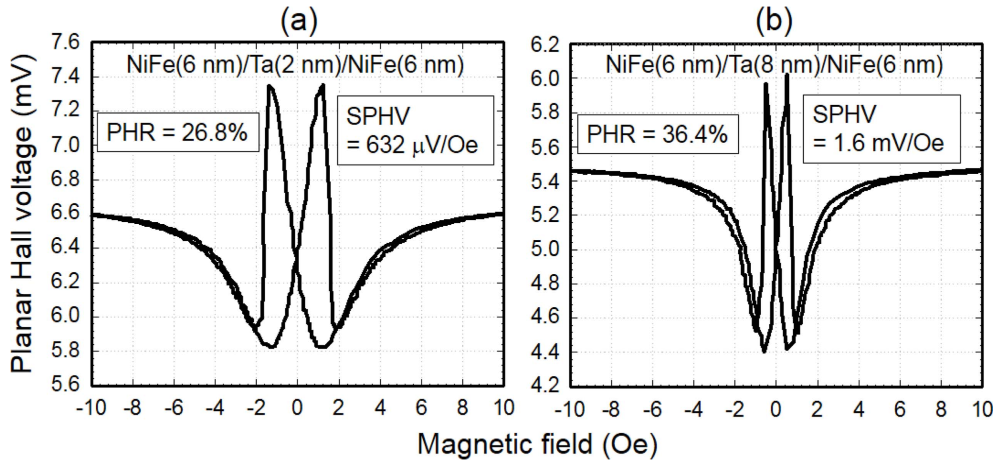

Fig. 3. PHV curves for the Ta(5 nm)/NiFe(6 nm)/Ta(t)/NiFe(6 nm)/Ta(5 nm) multilayer measured at the sensing current of 10 mA temperature in external magnetic fields of −10 Oe ~ +10 Oe; (a) t = 2 nm and (b) t = 8 nm. In the neighboring region of the external magnetic field of ± 1.0 Oe, the values of PHR ratio, MS, and SPHV are (a) 26.8 %, 11.2 %/Oe, 800 μV/Oe and (b) 36.4 %, 28 %/Oe, 1.6 mV/Oe, respectively.

was 90° to the easy axis, as shown in Fig. 1(b). The PHV curve was measured by the 4-terminal method, after adjusting the direction of the external magnetic field applied to the thin film sample at 90° to the direction of the voltage terminal along the horizontal plane of the thin film. Fig. 3(a) shows the PHV curve of the Ta(5 nm)/NiFe(6 nm)/Ta(2 nm)/NiFe(6 nm)/Ta(5 nm) multilayer sample with a sensing current of 10 mA. When the thickness of the ferromagnetic NiFe layer was 6 nm, the coercive force was 2.4 Oe. The coercive force shown in the PHV curves of Fig. 3(b) measured for Ta(5 nm)/NiFe(6 nm)/Ta(8 nm)/NiFe(6 nm)/Ta(5 nm) multilayer sample was equal to 1.3 Oe. The initial PHV and PHR values obtained from the same PHV curves in Fig. 3(a) and Fig. 3(b) as the magnetoresistive curve were 6.6 mV/5.45 mV and 0.65 Ω/0.545 Ω, respectively. The PHR ratio percentages were 26.8 % and 36.4 %, respectively. The PHV curves for these two types of thin film samples show a linearly high MS near the external magnetic field of 0 Oe. The MSs are 11.2 %/Oe and 28 %/Oe. The obtained SPHV for the Ta(5 nm)/NiFe(6 nm)/Ta(2 and 8 nm)/NiFe(6 nm)/Ta(5 nm) multilayer were 632 mV/Oe and 1.6 mV/Oe, respectively, indicating sufficient sensitivity to detect magnetic nanoparticle-antibody signals. As a result, it was shown that it can be used as a biosensor capable of measuring a fine magnetic field.

In addition, the dependence of the PHV, SPHV, PHR ratio, and MS on the thickness of Ta in the Ta/NiFe/Ta(t = 0, 2, 4, 8 nm)/NiFe/Ta multilayer is shown in Fig. 4. When the sensing current was 10 mA, the trends of PHV,

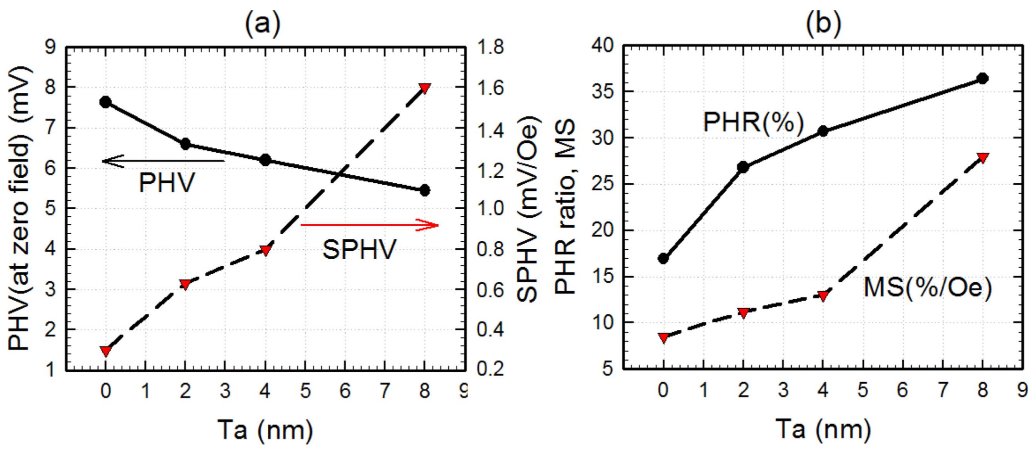

Fig. 4. (Color online) The dependence of (a) PHV and SPHV, (b) PHR ratio and MS for the thickness of Ta on the Ta(5 nm)/NiFe(6 nm)/Ta(t = 0, 2, 4, 8 nm)/NiFe(6 nm)/Ta(5 nm) multilayer. Here SPHV (mV/Oe), PHR ratio (%), and MS (%/Oe) obtained by the PHV curves in the region of applied magnetic field of –2.0 Oe ~ +2.0 Oe.

SPHV, and PHR at 0 Oe decreased, increased, and decreased with an increase in the thickness of Ta, respectively. As a result, the SPHV increased from 280 μV/Oe to 1.6 mV/Oe as the Ta thickness value increased from 0 nm to 8 nm. When the sensing current increased to 40 mA, PHV and SPHV of the Ta(5 nm)/NiFe(6 nm)/Ta(4 nm)/NiFe(6 nm)/Ta(5 nm) multilayer increased to 6.4 mV and 3.2 mV/Oe, as shown in Fig. 2. Alternatively, when the sensing current increased from 1 mA to 40 mA, the PHR caused by Joule's heat generated at the intersection point decreased from 30.7 % to 28.5 %. From these results, in a GMR-SV thin film with a dual structure, the interlayer coupling between the free layer and the pinned layer is minimized, and the thickness of the Cu layer inserted to control the exchange of coupling field between the antiferromagnetic layer and the pinned ferromagnetic layer. It can be used as a PHR biosensor, which is much better with a thin film having a double multilayered structure composed of only NiFe and Ta thin films, in addition to being a possible method to enhance the sharpened linear property determining the SPHV.

Magnetic nanoparticles are being commercialized by introducing them into various biochemical separation and purification techniques. Wang *et al.* constructed a magnetic nanosensor in the form of an enzyme-linked immunospecific assay (ELISA), in the antigen measurement method after combining the antibody and nanoparticles [15, 16]. Unlike the fluorescence and absorption methods that are affected by the surrounding environment, it was used in an ultra-sensitive analysis device that is not affected by any environmental parameter such as pH, temperature, ionic strength, and self-luminescence. It is used in biomarker measurement technology [15]. Recently, magnetic nanoparticles with a core-shell structure composed of Fe_3O_4 and zinc oxide (ZnO) were mounted on dendritic cells, allowing magnetic resonance imaging (MRI) to monitor where dendritic cells are delivered in the human body. In addition, it has been reported by local researchers that magnetic nanoparticles can significantly improve the delivery efficiency of dendritic cells with the help of the tumor-specific antigens on the surface of magnetic nanoparticles [17].

4. Conclusion

In this study, the voltage properties of a planar Hall device with a Ta/NiFe/Ta/NiFe/Ta multilayer that was prepared in a cross-shaped pattern at a width of 0.5 mm using a shadow mask were investigated according to the thickness of the inserted Ta layers. The SPHV obtained from the PHV curves of the Ta(5 nm)/NiFe(6 nm)/Ta(t = 0-8 nm)/NiFe(6 nm)/Ta(5 nm) multilayered sample was a 0.3-1.6 mV/Oe in the fine magnetic field region of the external magnetic field with ± 1.0 Oe. When the thickness of the inserted Ta layer was 4 nm, the PHR ratio, MS, and SPHV were 30.7 %, 13 %/Oe, and 800 mV/Oe, respectively. In particular, at a 8 mm thickness of Ta, the PHR ratio, MS, and SPHV improved significantly by as much as 36.4 %, 28 %/Oe, and 1.6 mV/Oe, respectively. These results can be used in the development of a PHR biosensor with a high sensitivity for the detection of nanoparticle-antibody binding, even in a simple structure formed only out of a ferromagnetic NiFe layer.

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