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Magnetostrictive GMR spin valves with composite FeGa/FeCo free layers

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We have fabricated strain-sensitive spin valves on flexible substrates by utilizing the large magnetostrictive FeGa alloy to promote the strain sensitivity and the composite free layer of FeGa/FeCo to avoid the drastic reduction of giant magnetoresistance (GMR) ratio. This kind of spin valve (SV-FeGa/FeCo) displays a MR ratio about 5.9%, which is comparable to that of the conventional spin valve (SV-FeCo) with a single FeCo free layer. Different from the previously reported works on magnetostrictive spin valves, the SV-FeGa/FeCo displays an asymmetric strain dependent GMR behavior. Upon increasing the lateral strain, the MR ratio for the ascending branch decreases more quickly than that for the descending branch, which is ascribed to the formation of a spiraling spin structure around the FeGa/FeCo interface under the combined influences of both magnetic field and mechanical strain. A strain sensitivity of GF = 7.2 was achieved at a magnetic bias field of -30 Oe in flexible SV-FeGa/FeCo, which is significantly larger than that of SV-FeCo. [http://dx.doi.org/10.1063/1.4943770]

Spin-valve structures based on the giant magnetoresistance (GMR) effect,[1,2] typically including a ferromagnetic (FM) free layer and an FM/antiferromagnetic (AFM) exchange biased bilayer separated by a nonmagnetic (NM) conductive layer,[3] have been broadly applied in magnetic field sensors with high sensitivity.[4,5] The FM free layers with a small magnetostrictive coefficient, such as $Fe_{10}Co_{90}$ (~ 3 ppm),[6] are often required to reduce the magnetic anisotropy generated by stress during the preparation and therefore to ensure the high magnetic field sensitivity. On the other hand, if a high magnetostrictive material is utilized as the free layer in a spin valve, external strain can effectively change the magnetization orientation of the free layer by means of the inverse magnetostrictive effect, leading to a remarkable change in magnetoresistance.[7–18] Such magnetostrictive spin valves, displaying the coupled magnetostrictive–magnetoresistive behaviors, can be designed as novel strain sensors with the advantages of small size, high sensitivity, and easy to integrate in a digital circuit.[19] The strain sensitivity for a GMR strain sensor is usually defined as the gauge factor $GF = (\Delta R/R)/\Delta\varepsilon$, where ΔR is the resistance change when varying the external strain by $\Delta\varepsilon$. Dueuas *et al*. demonstrated that the strain dependent MR effect can be significantly enhanced by using an optimized magnetostrictive $Fe_{50}Co_{50}$ (~ 100 ppm) as the free layer of spin valves.[8,20] Löhndof *et al*. introduced the $Fe_{50}Co_{50}$ free layer into magnetic tunnel junction (MTJ) structures prepared on rigid substrates and achieved a very high strain sensitivity on the order of 600.[11] Since the Young's modulus of a stiff and thick substrate such as Si wafer is one or two order higher than that of flexible substrate such as polymers, the operational range for a magnetostrictive spin-valve

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stack grown on Si is by far less than that prepared on flexible substrates, but the corresponding strain sensitivity for flexible spin valves is remarkable reduced as compared to the rigid ones. For instance, Uhrmann *et al*. obtained an enhanced operational range of 3% but a reduced gauge factor of 2.2 in flexible magnetostrictive GMR structures prepared on polyimides.[13]

For a practical strain sensor based on the GMR effect, a high MR ratio and a magnetostrictive sensing layer are essential to improve the response to external strain. It is well known that $Fe_{81}Ga_{19}$ (FeGa) alloys exhibit the largest magnetostriction (~350 ppm for the typical bulk) among the various alloys not containing rare earth elements.[21] For FeGa alloys used as the sensing layer of spin valves, the large magnetostriction is able to improve the response of GMR behaviors to applied strain. However, the utilization of FeGa in spin valves may result in the obvious reduction of MR ratio, which trades off the advantage brought by the large magnetostriction. In previous works, inserting a thin $Fe_{10}Co_{90}$ (FeCo) or Co layer adjacent to the NM conductive layer can dramatically increase the spin-dependent scatting of polarized conduction electrons at the interface and enhance the MR ratio of spin valves, providing an important strategy for building magnetic sensors with a high magnetic field sensitivity.[22,23] In this work, we fabricated flexible GMR spin valves (SV-FeGa/FeCo) with the composite free layers of FeGa/FeCo. As compared to the conventional spin-valve structures (SV-FeCo), the composite free layer can improve the response of GMR behaviors to the external strain and meanwhile avoid the drastic reduction of MR ratio in SV-FeGa/FeCo. In addition, this kind of flexible spin valves displays a distinct strain dependent GMR behavior, that is, with straining the MR ratio for the descending branch reduces much slower than that for the ascending branch, which can be interpreted by considering the formation of a spiraling spin structure at the FeGa/FeCo interface under the combined influences of both magnetic field and applied strain.

Spin-valve structures with multilayers of Ta(4.5nm)/free layers/Cu(3 nm)/FeCo(5 nm)/IrMn(15 nm)/Ta(3 nm), as schematically displayed in Fig. 1(a), were prepared on flexible polyethylene terephthalate (PET) substrates by using a magnetron sputtering system with a base pressure of about 5.0×10^{-8} mTorr. The 3 nm Cu spacer layer has been demonstrated to be thick enough to magnetically decouple the free and pinned layers. Before transferred into the vacuum chamber, the PET plastics were spin-coated with a photoresist layer to reduce the roughness. For comparison, the composite free layer of FeGa(2 nm)/FeCo(2 nm) and the single free layer of FeCo(4 nm) were used in the samples of SV-FeGa/FeCo and SV-FeCo, respectively. An in-plane magnetic field of 500 Oe provided by a permanent magnet was applied during deposition to induce the exchange bias. In order to avoid a considerable magnetic anisotropy induced by the internal strain and the oblique deposition in free layers, we grew the multilayered spin valves at room temperature and with rotating the substrates. The magneto-transport characterizations for the spin valves under different applied strains were conducted at room temperature with a standard four-point probe setup installed on a home-designed stretching apparatus. Both the electric current and the magnetic field were applied

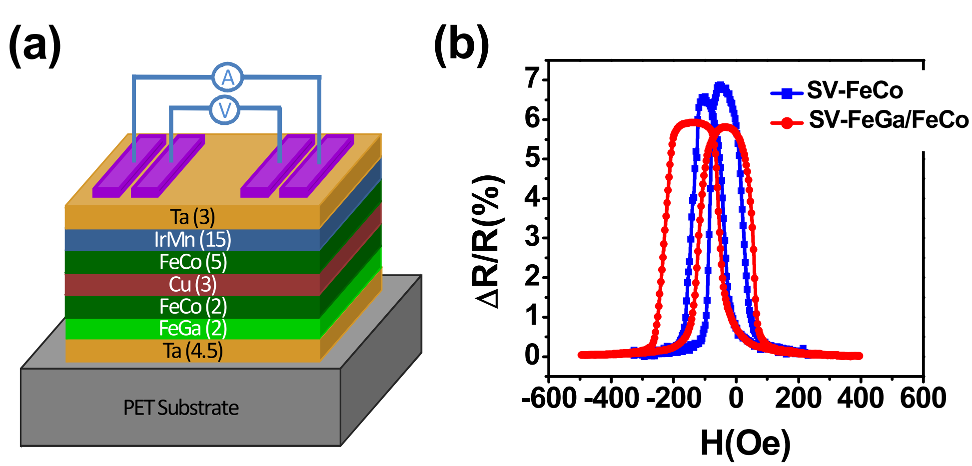

FIG. 1. (a) Schematic diagram of multilayer stack of flexible SV-FeGa/FeCo with the composite free layers. (b) GMR curves of the as-fabricated SV- FeCo and SV-FeGa/FeCo.

along the exchange bias. The mechanical tensile strain in a precise of 0.2% can be applied parallel or perpendicular to the exchange bias.

The flexible SV-FeCo display an MR ratio of 6.9%, as illustrated in Fig. 1(b), which is slightly lower than this multilayer stack grown on rigid substrates due to the rough surface of PET membranes, although the root-mean-square roughness of PET has been reduced from 1.67 to 0.48 nm by means of spin-coating a photoresist layer. It should be noted that some other solutions of organic materials, such as PET, polydimethylsiloxane (PDMS), polyimide (PI), can replace photoresist to be spin-coated on flexible substrate to decrease the surface roughness. After a magnetostrictive FeGa layer is added into the free layer, the MR ratio of SV-FeGa/FeCo with the composite free layers decreases to 5.9%. Because of the dominant role of the FeCo/Cu interfacial scattering in determining the GMR behaviors, the addition of magnetostrictive FeGa layer does not lead to an obvious decrease in MR ratio, but the reduced thickness of FeCo layer is responsible for the slight reduction of MR ratio. In contrast, if using a single FeGa free layer but not the composite free layer, the MR ratio will be significantly reduced to about 1.7%.

Figure 2(a) displays the GMR behaviors for the conventional SV-FeCo with a tensile strain applied perpendicular to the exchange bias of spin valves. When the lateral strain increases from 0 to 1.8%, the MR ratio of SV-FeCo significantly decreases from 6.9% to 5.7%. Correspondingly, the loop squarenesses of free layer and pinned layer are reduced from 0.79 to 0.37 and 0.87 to 0.84, respectively. It is well known that the uniaxial magnetic anisotropies for the grains in polycrystalline magnetic films do not strictly orient along in an identical direction, but have a distribution around their average direction. The transverse strain can decrease the strength of uniaxial magnetic anisotropy of FM layer and induce a broad distribution of magnetic anisotropies.[24–26] As a result, the orientations of the magnetization in the free and pinned layers become deviated from the antiparallel alignment with rising strain, which leads to the decrease of both MR ratio and loop squareness.

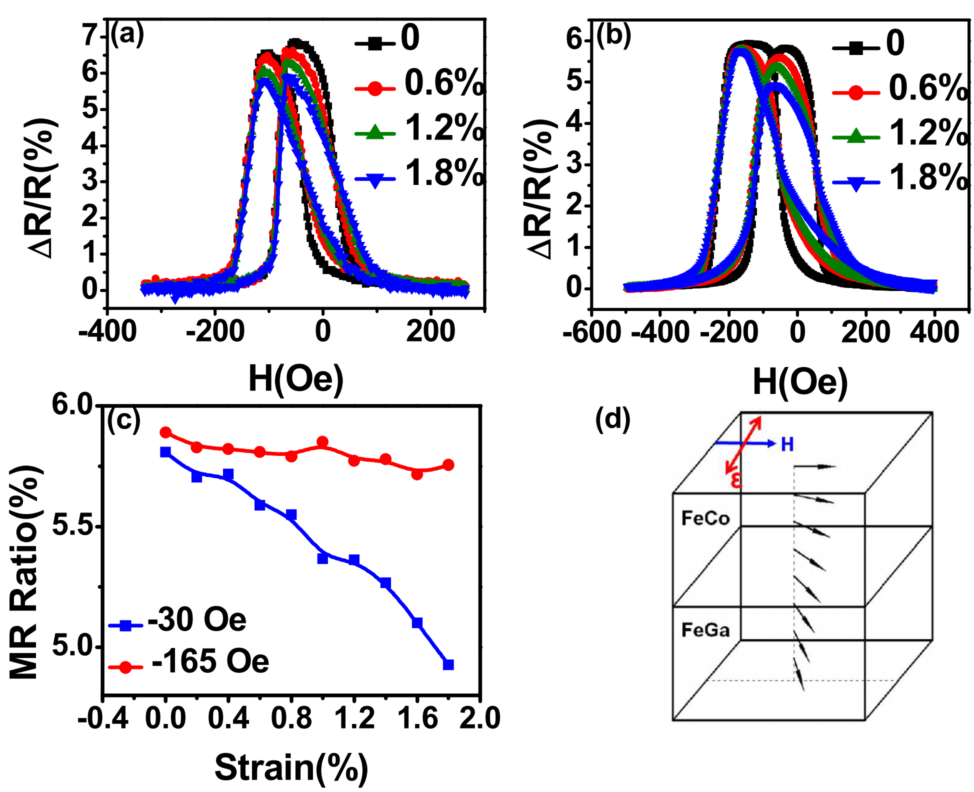

FIG. 2. GMR curves for (a) SV-FeCo and (b) SV-FeGa/FeCo under different lateral strains. (c)The lateral strain dependence of MR ratio for the ascending branch (measured at -30 Oe) and the descending branch (measured at -165 Oe) in SV-FeGa/FeCo. (d) Schematic diagram of the spiraling spin structure occurring around the FeGa/FeCo interface under the combined influences of both magnetic field and mechanical strain.

After releasing the applied strain, the MR ratio can be restored to the original value of 6.9%. For the conventional SV-FeCo, the two high-resistance states occurring with ascending and descending magnetic field are located around zero magnetic field and the exchange bias field, respectively. They display the same strain dependent behaviors. In contrast, upon increasing the lateral strain on the magnetostrictive SV-FeGa/FeCo, the MR ratio for the descending branch (measured at -165 Oe) displays a slight reduction from 5.9% to 5.8% with the applied strain increasing from 0 to 1.8%, but the corresponding MR ratio for the ascending branch (measured at -30 Oe) decreases much quickly from 5.9% to 4.9%, as revealed in Figs. 2(b) and 2(c). The distinct strain dependent GMR behaviors of SV-FeGa/FeCo can be ascribed to the formation of a spiraling spin structure around the FeGa/FeCo interface under the combined influences of both magnetic field and lateral strain, as schematically shown in Fig. 2(d).[27,28] Due to the inverse magnetostrictive effect, the magnetic moments of FeGa are preferentially aligned along the lateral tensile strain. The interfacial exchange coupling between FeGa and FeCo drives the FeCo moments towards to the tensile strain. On the other hand, for the descending branch, the considerable magnetic field forces the magnetic moments of FeCo parallel to the longitudinal direction. Consequently, the orthogonal configuration of magnetic field and applied strain used in the magneto-transport measurement leads to the formation of spiraling spin structure. In this scenario, the FeCo moments in both FM layer, especially close to the FeCo/Cu interface, can form the rather good parallel or antiparallel alignment even when raising the external strain. Therefore, the MR ratio does not significantly decrease with straining. However, for the ascending branch, the antiparallel magnetic configuration appears around zero magnetic field. The relatively low magnetic field cannot result in the formation of spiraling spin structure, so the MR ratio decreases rapidly with straining, which is similar to the case of SV-FeCo. It should be noted that when a tensile strain up to 1.8% is applied parallel to the exchange bias of spin valves, only a slight increase of MR ratio can be observed in both SV-FeCo and SV-FeGa/FeCo. This can be understood that the longitudinal strain can increase the strength of uniaxial magnetic anisotropy of FM layer, squeeze the orientations of magnetic anisotropies into a narrow distribution, and therefore slightly enhance the MR ratio.

For a strain sensor based on the GMR effect in practical application, it is hard to obtain the GMR ratio by applying a variable magnetic field. The feasible way is to measure the change in resistance when applying an external strain. In addition, a fixed magnetic bias field need to be applied to obtain the high-resistance state R_{AP} occurring at the antiparallel magnetic configuration, thus to achieve a good response of resistance to applied strain.[10,16] Figure 3(a) shows the sample resistance as a function of applied strain for the both spin valves. The resistances for SV-FeCo and SV-FeGa/FeCo are respectively measured with ascending the magnetic field at -50 Oe and -30 Oe where the resistances show the maximum values. With raising the lateral strain up to 1.8%, the resistances for SV-FeCo and SV-FeGa/FeCo vary from 1.99 to 2.01 Ω and from 8.85 to 10.0 Ω, respectively. Thus, the strain sensitivities for SV-FeCo and SV-FeGa/FeCo can be obtain as GF

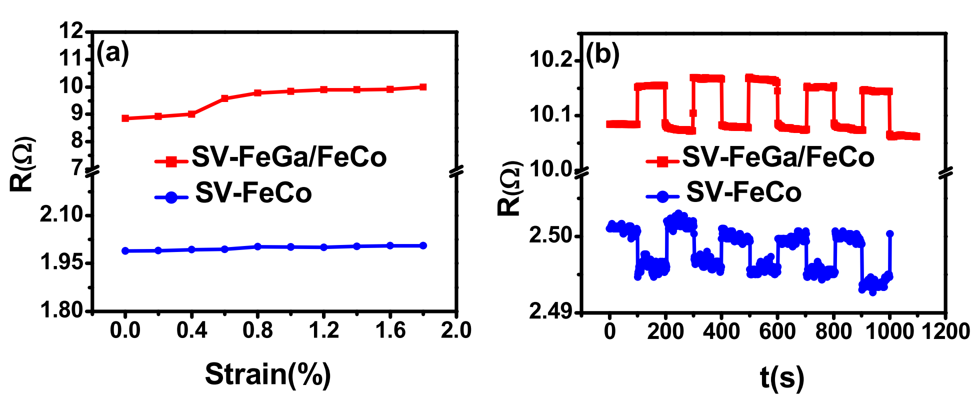

FIG. 3. (a) The lateral strain dependence of sample resistance for SV-FeCo and SV-FeGa/FeCo measured with ascending the magnetic field at -50 Oe and -30 Oe, respectively. (b)The variation of resistance for both SV-FeCo (measured at a bias field of -50 Oe) and SV-FeGa/FeCo (measured at-30 Oe) under a sequential cyclic stretching test with a stretch loading of 0.6%.

= 0.56 and 7.2, respectively. As compared to SV-FeCo, the remarkable enhancement of GF in SV-FeGa/FeCo can be ascribed to the addition of the magnetostrictive FeGa layer into the free layer. However, the strain dependent resistance for spin-valves does not display good linearity within the whole measurement range. Only for the strain below 0.4% for SV-FeGa/FeCo and below 0.6% for SV-FeCo, the linearity of resistance-stain effect is rather good. In contrast, the linearity for the GMR/TMR based strain sensors grown on Si is usually better than that of our flexible ones.[10,16] This is because of the complicated strain status in flexible substrates, which induces a distribution of magnetic anisotropy in magnetic layers. Consequently, when the application of strain changes the easy axis to the hard axis, the hysteresis loop cannot change from a perfect rectangle with a squareness of one to a sheared one with the remanence of zero, but displays a continuous change in squareness.[24,25] Another reason is that due to the difference in elastic parameters, the strain applied on rigid Si is usually very small (less than 0.1%),[10,16] which is by far less than the stain (~1.8%) we applied on the flexible spin valves. It should be noted that during the stretching measurement, the low-resistance state occurring at the parallel magnetic configuration shows a small increase when increasing the strain due to the elastic deformation of metallic films. In order to assess the stretching fatigue of the magnetostrictive spin valves, we conducted a sequential cyclic stretching test with a stretch loading of 0.6%. Figure 3(b) shows that a high resistance for both SV-FeCo (measured at a bias field of -50 Oe) and SV-FeGa/FeCo (measured at -30 Oe) appears at the external strain of 0.6%. After removing the strain, the sample resistances can be roughly recovered to the original values, displaying a good repeatable behavior under a cyclic loading test. The fluctuation of the high and low resistance states is arisen from the slight deformation of flexible substrates after cyclic stretching, because our home-designed stretching apparatus could not provide a perfect uniform tensile strain across the whole sample.

In summary, we designed and fabricated a magnetostrictive spin valve with a composite FeGa/FeCo free layer on flexible PET plastics. The SV-FeGa/FeCo displays a MR ratio of 5.9%, which is comparable to that of the conventional SV-FeCo. Due to the combined effects of both magnetic field and mechanical strain, a spiraling spin structure was found around the FeGa/FeCo interface. Consequently, the SV-FeGa/FeCo exhibits a distinct strain dependent GMR behavior. When applying a lateral strain, the MR ratio for the descending branch reduces much slower than that for the ascending branch. A strain sensitivity of GF = 7.2 was achieved in flexible SV-FeGa/FeCo, which is significantly larger than that of SV-FeCo.

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