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Influence of thermal deformation on exchange bias in FeGa/IrMn bilayers grown on flexible polyvinylidene fluoride membranes

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We investigated the influence of thermal deformation on the magnetic properties of flexible FeGa/IrMn exchange biased (EB) bilayers grown on polyvinylidene fluoride (PVDF) membranes with the pinning direction parallel to the thermal deformation directions α_{31} and α_{32} of PVDF. Because with increasing temperature the uniaxial anisotropy is reoriented from the α_{32} to α_{31} directions by the thermally induced compressive strain along the α_{32} direction, both the coercive field H_c and the loop squareness M_r/M_s of FeGa/IrMn bilayers display a complex temperature dependent behavior. The EB field H_c decreases with increasing temperature. A modified Stoner-Wohlfarth model was employed to account for the temperature dependence of magnetic properties in the flexible FeGa/IrMn bilayers.

***Index Terms*—flexible magnetoelectronics, exchange bias, temperature modulation, thermal expansion**

I. Introduction

Exchange bias (EB) refers to the interfacial exchange coupling between ferromagnetic and antiferromagnetic layers and has been widely applied in magnetoelectronic devices such as magnetic tunneling junctions (MTJ) and spin-valve sensors [1-5]. Nowadays, the EB related flexible magnetic multilayers have found their application in wearable electronics, such as curved MTJ and flexible spin valve magnetic sensors, which are stretchable and bendable, and hence are closely strain related [6-9]. The most common strain in flexible devices is the thermal expansion or contraction, especially in plastic substrates with large thermal expansion coefficient. Thermal-strain-induced magnetic anisotropy could change the magnetization orientation of the EB layers, thus destabilizing the magnetotransport properties of devices [9].

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Therefore, the investigation on the effect of thermal deformation on the magnetic properties of EB bilayers is of great significance for the development of flexible magnetoelectronic devices. The effect of mechanical strain on magnetic properties for EB systems grown on rigid inorganic substrates, including NiFe/NiO, NiFe/IrMn, and $Fe/Fe_{0.6}Zn_{0.4}F_2(110)$ have been studied previously [10-12]. However, the maximum strain in the order of 0.1% can be achieved in the stiff substrates, which is much lower than the thermal deformation in flexible systems. In the past few years, we reported that the magnetic anisotropy in flexible magnetic films can be effectively modulated by a bending strain in the order of 1% [9, 13]. The flexible polyvinylidene fluoride (PVDF) membrane possesses a large anisotropic negative thermal expansion coefficient of $\alpha_{31} = -13$ ppm/K, and $\alpha_{32} = -145$ ppm/K [14, 15]. By changing the temperature, a large anisotropic thermal strain could be generated in the PVDF substrate, which can be effectively transferred to the EB heterostructures grown it.

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In this study, we investigated the effect of anisotropic thermal deformation on the magnetic properties of flexible $Fe_{81}Ga_{19}/Ir_{20}Mn_{80}$ bilayers grown on PVDF substrates. The magnetostrictive $Fe_{81}Ga_{19}$ alloy which exhibits a large magnetostriction coefficient (~350 ppm for the typical bulk) is selected as the ferromagnetic layers to improve the response of magnetic properties to the mechanical strain induced by the thermal deformation of PVDF. The coercivity (H_c), EB field (H_{eb}), and loop squareness (M_r/M_s) of FeGa/IrMn heterostructures can be effectively changed by the thermal strain. A simple simulation was conducted to well understand the thermal deformation control of magnetic properties in the flexible EB system.

II. Experiment

The samples with a layer structure (from the top down) of Ta(3 nm)/$Fe_{81}Ga_{19}$(10 nm)/$Ir_{20}Mn_{80}$(20 nm)/Ta(20 nm) were deposited on flexible PVDF substrates by using a magnetron sputtering system with a base pressure below 3×10^{-7} torr at 290 and 310 K. A reference sample was fabricated on rigid Si wafers at 290 K. FeGa thin films were polycrystalline and mainly were textured with the (110) direction out of plane due to presence of pseudo-orthorhombic symmetry in the bcc structure [16, 17]. The bottom Ta seeding layers were employed to reduce the roughness of PVDF membranes and to induce the (111) texture growth of IrMn layer which were enhanced when Ta got a strongest (200) β–Ta texture [18]. A magnetic field, H_{growth} = 800 Oe, provided by a permanent magnet, was applied along the α_{31} and α_{32} directions of PVDF during deposition to induce an EB. To study the effect of thermal deformation of PVDF on the magnetic properties of FeGa/IrMn EB heterostructures, the hysteresis loops were measured at various temperatures by Quantum Design physics property measurement system. The magnetic field was cycled ten times to minimize the training effect. Subsequently, the hysteresis loops were measured from 200 to 350 K in an interval of 30 K.

III. Results and Discussion

Figure 1 shows the hysteresis loops of the FeGa/IrMn EB bilayers deposited on both Si and PVDF substrates at 290 K. As shown in Figs. 1(a) and 1(b), the sample on rigid Si substrate displays a maximum H_{eb} = 88.7 Oe and H_c = 78.7 Oe at 200 K along the pinning direction (PD, θ = 0°). With increasing the temperature, H_{eb} and H_c decrease monotonously. For the magnetic field applied perpendicular to the PD (θ = 90°), H_{eb} is around 0 Oe and H_c keeps at 6.7 Oe in the temperature range from 200 to 350 K. For flexible FeGa/IrMn bilayer grown on PVDF membrane with the EB set along the α_{31} direction, the hysteresis loops with the magnetic field applied parallel and perpendicular to the PD are shown in Figs. 1(c) and 1(d), respectively. For the scenario of EB along the α_{32} direction, the hysteresis loops measured along and perpendicular to the PD are displayed in Figs. 1(e) and 1(f), respectively. The configurations of measurement are schematically illustrated in the insets of Figs. 1(c) and 1(e). Because of the large anisotropic thermal deformation of PVDF, the effect of temperature on H_c, H_{eb}, and loop squareness are quite evident. The values of H_c, H_{eb}, and M_r/M_s ratio for the samples measured along and perpendicular to the PD are summarized in Fig. 2.

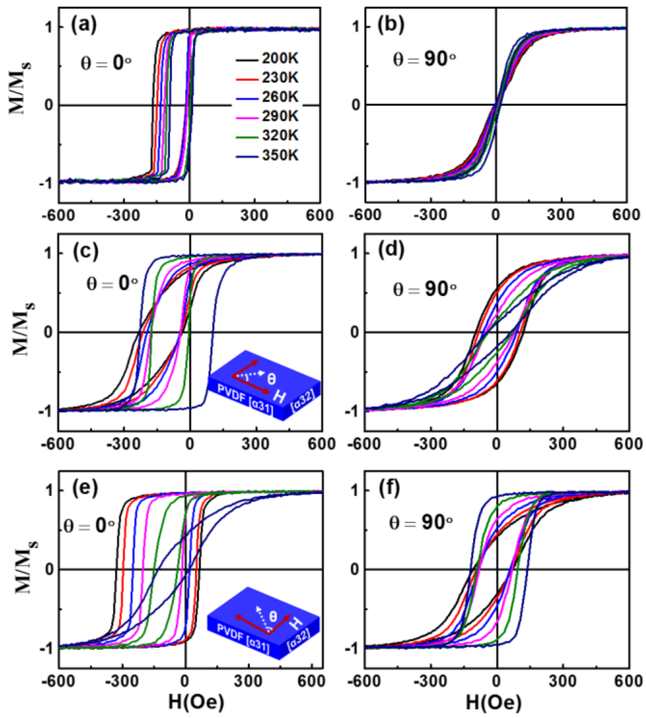

Fig. 1. Hysteresis loops measured at different temperatures for the FeGa/IrMn bilayers deposited on (a, b) rigid Si substrates, and on flexible PVDF membranes with the PD set (c, d) along the α_{31} direction and (e, f) along the α_{32} direction. The magnetic field is applied at (a, c, e) θ = 0° and (b, d, f) θ = 90° with respect to the PD. The configurations of measurement are schematically indicated in the insets of (c) and (e), respectively.

Considering the thermal expansion coefficient α_{32} is larger than α_{31}, the expansion or contraction of PVDF membranes driven by the difference between the measuring temperature and the deposition temperature will exert an in-plane strain on the magnetic layers, leading to the

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change of magnetic properties due to the inverse magnetostrictive effect. When the measuring temperature is lower (higher) than the growth temperature, a tensile (compressive) strain is induced along the α_{32} direction. It should be noted that the strain cannot be fully transferred to the magnetic layers because of the appearance of cracks in the films and the large difference of the Young's moduli between the metallic films and the flexible substrates [19]. Here we define the strain transfer efficiency, η, which will be discussed below.

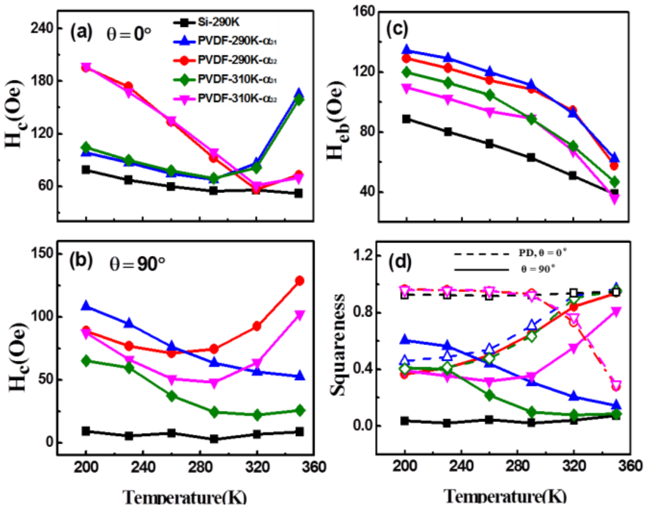

Fig. 2. Temperature dependence of H_c measured (a) along the PD and (b) perpendicular to the PD. Temperature dependence of (c) H_{eb} and (d) loop squareness, solid line and dashed line correspond to the measurement along the PD and perpendicular to the PD, respectively. The FeGa/IrMn samples are deposited on Si at 290 K and on PVDF at both 290 and 310 K with the PD set along both the α_{31} and α_{32} directions.

Figure 2(a) shows when temperature increases from 200 to 350 K, H_c for FeGa/IrMn bilayer grown on Si substrate and measured at $\theta = 0°$ decreases from 78.7 Oe to 51.8 Oe due to the reduced interfacial exchange coupling and random field caused by thermal agitation [20, 21]. For the sample with the PD parallel to the α_{31} direction, H_c at $\theta = 0°$ shows a minimum at 290 K. For the samples grown on PVDF, when the PD is parallel to the α_{32} direction, H_c measured at $\theta = 0°$ appears a minimum at 320 K because with increasing temperature the uniaxial anisotropy is reoriented from the α_{32} to α_{31} directions under a thermally induced compressive strain along the α_{32} direction. As shown in Fig. 2(b), H_c measured at $\theta = 90°$ keeps unchanged around 7 Oe for the sample deposited on Si. For the sample grown on PVDF with the PD along the α_{31} direction, H_c at $\theta = 90°$ decreases from 108.1 to 52.6 Oe and from 65.0 to 25.8 Oe when the films are deposited at 290 and 310 K, respectively. As for the sample with the PD parallel to the α_{32} direction, H_c at $\theta = 90°$ shows a minimum around 260 K. As shown in Fig. 2(c), with temperature increasing from 200 to 350 K, H_{eb} for the sample deposited on Si decreases linearly from 88.7 to 38.7 Oe. The similar decrease of H_{eb} can be found for the samples deposited on PVDF in the temperature range from 200 to 290 K, which suggests that the thermal agitation is the main factor to decrease H_{eb}. When the measuring temperature is higher than 290 K, the decrease of H_{eb} becomes steeper, indicating that the compressive strain generated the thermal deformation of PVDF could accelerate the decrease of H_{eb}. Figure 2(d) shows the loop squarenesses (M_r/M_s) obtained both along the PD (dashed line) and perpendicular to the PD (solid line). The intersection point of the dashed line and solid line for a sample corresponds to the critical temperature where the FeGa layer reaches magnetically isotropic. The loop squareness for the sample on Si remains 0.93 and 0.05 with the magnetic field applied along and perpendicular to the PD, respectively, indicating that the thermal agitation has a negligible effect on the loop squareness. For the flexible samples on PVDF with the PD set along the α_{32} direction, regardless of the deposition temperature, the squareness obtained at $\theta = 0°$ decreases from 0.96 to 0.28 with increasing temperature from 200 to 350 K. The uniaxial anisotropy is switched from the α_{32} to α_{31} directions with increasing temperature. For the case of $\theta = 90°$, the squareness increases from 0.37 to 0.94 and from 0.39 to 0.82 for the flexible samples deposited at 290 and 310 K, respectively. The corresponding critical temperatures of magnetic isotropy are around 311 and 329 K. Since the EB induces an additional uniaxial magnetic anisotropy along the α_{32} direction, in order to achieve the magnetic isotropy in the FeGa layers, a collinear compressive strain needs to be produced through the anisotropic thermal deformation of PVDF substrate with increasing the temperature slightly beyond the growth temperature. The thermal strain may induce an orthogonal uniaxial anisotropy to counteract the EB-induced uniaxial anisotropy. As for the flexible samples deposited both at 290 and 310 K with the PD parallel to the α_{31} direction, the squareness obtained at $\theta = 0°$ increases from 0.45 to 0.95 with increasing the temperature from 200 to 350 K. For $\theta = 90°$, the squareness decreases from 0.61 to 0.15 and from 0.41 to 0.08 for the samples deposited at 290 and 310 K, respectively. The corresponding critical

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temperatures of magnetic isotropy appear at 243 and 211 K. In this case, the EB-induced uniaxial magnetic anisotropy in the FeGa layer is along the α_{31} direction. Therefore, a tensile strain along the α_{32} direction needs to be produced with decreasing the temperature below the growth temperature, so that the FeGa layer could reach magnetically isotropic.

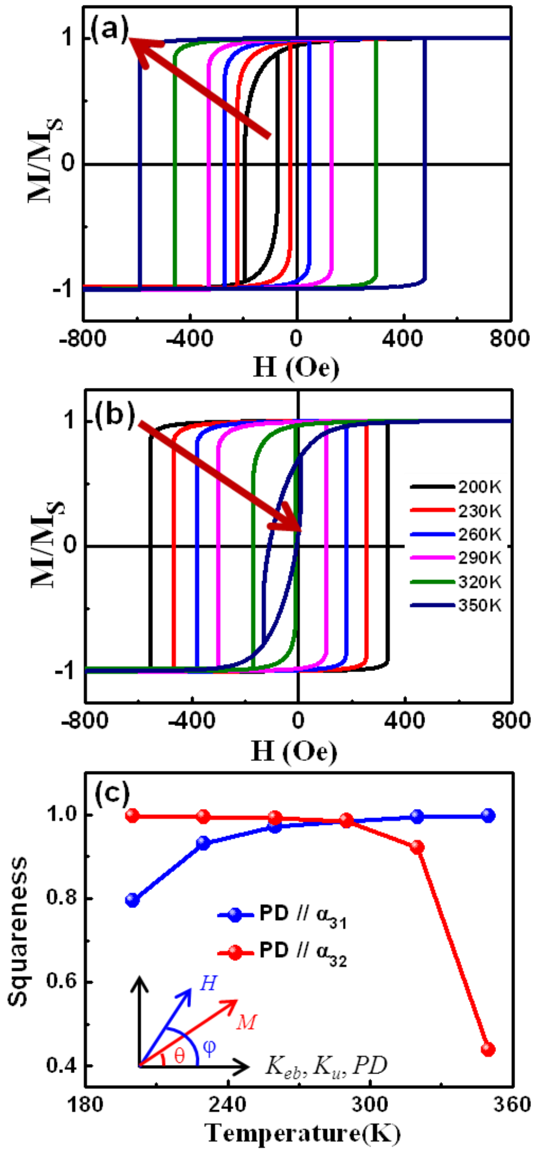

Fig. 3. Calculated hysteresis loops at various temperatures for flexible FeGa/IrMn bilayers on PVDF with the PD (a) along the α_{31} and direction and (b) along the α_{32} direction. The corresponding temperature dependence of loop squareness is plotted in (c).

In order to explain the effect of measuring temperature on the magnetic properties of flexible FeGa/IrMn EB bilayers, we carried out a simple simulation based on the Stoner-Wohlfarth model [22]. The total free energy of an EB system can be written as:

$$E = -K_u \cos^2\varphi + K_\sigma \cos^2\varphi - K_{eb}\cos\varphi - M_s H\cos(\varphi-\theta),$$

where M_s = 833 emu/cm^3 is the saturation magnetization, H is the applied magnetic field, θ is the angle between H and PD direction, φ is the angle between magnetization and magnetic easy axis, K_u is the EB-induced uniaxial anisotropy, K_σ is the thermal-strain-induced uniaxial anisotropy, K_{eb} is the EB-induced unidirectional anisotropy. K_u is given by $K_u = 1/2H_kM_s$. K_{eb} is given by $K_{eb} = H_{eb}M_s$. In this experiment, the strain is generated by the anisotropic thermal deformation of PVDF by changing the temperature. Considering the strain transfer efficiency between the magnetic films and the PVDF membranes, the strain actually exerted on the films is $\varepsilon_F = \eta(\alpha_{31} - \alpha_{32})(T - T_0)$, where T and T_0 represent the measuring temperature and the deposition temperature, respectively. Consequently, K_σ can be expressed as:

$$K_\sigma = -\frac{3}{2}\lambda_s\sigma = -\frac{3}{2}\lambda_s\eta(\alpha_{31}-\alpha_{32})(T-T_0)E_f/(1-\nu^2),$$

where E_f = 60 GPa is the Young's modulus of FeGa, λ_s = 60 ppm is the saturation magnetostriction constant of FeGa films [9], η = 20%, and ν = 0.3 is the Poisson ratio for metals. It is reported that the thermal expansion coefficients of PVDF are varied with temperature [23]. The value of α_{32} and α_{31} increases with increasing temperature. Therefore, in order to achieve the best fitting loops, the different thermal expansion coefficients at different temperatures (for example, α_{31}–α_{32}= 78 ppm/K for 200 K, α_{31}–α_{32}= 132 ppm/K for 290 K) are used for our numerical calculations [23]. In simulation, K_{eb}/M_s acquired through the experimental H_{eb} measured at different temperatures and K_u/M_s = 150 Oe are used. Accordingly, based on the magnetization reversal mechanism of coherent rotation, the calculated hysteresis loops are obtained at various temperatures for the flexible FeGa/IrMn. The growth temperature is set at T_0 = 290 K. Figures 3(a) and 3(b) show the calculated hysteresis loops for the PD parallel to the α_{31} and α_{32} directions, respectively. The value of H_c increases with increasing temperature for the PD along the α_{31} direction, but decreases with temperature when the PD is parallel to the α_{32} direction. This variation of H_c is consistent with our experiment, as shown in Fig 2(a). It is worth noting that the calculated H_c are much bigger than the experimental values because the magnetization reversal mechanism should partially involve domain wall motion under low magnetic field, but our theoretical model is merely based on the magnetization rotation. Figure 3(c) shows the loop squareness as a function of temperature for the PD along the α_{31} and α_{32} directions. When the PD is parallel to the α_{31} direction, the loop squareness increases from 0.79 to 1.0 with increasing the temperature from 200 to 350 K. However, for the PD along the α_{32} direction, the loop squareness reduces from 1.0 to 0.36. The change of squareness with temperature is well consistent with our experimental observations, which can be mainly attributed to the changes of the strain-induced uniaxial magnetic anisotropy due to the large anisotropic thermal deformation of PVDF. When the measuring temperature is lower (higher) than the growth temperature, a tensile (compressive) strain is induced along the α32 direction. As a result, the thermally induced uniaxial anisotropy changes from the α32 to α31 directions when the temperature changes from lower to higher than the growth temperature. Since there is another uniaxial anisotropy induced exchange bias along the α31 and α32 directions, the total uniaxial anisotropy changes its direction depending on the relative magnitude between the thermally induced uniaxial anisotropy and the EB-induced uniaxial anisotropy.

IV. Summary

In conclusion, we fabricated FeGa/IrMn heterostructures on Si and flexible PVDF substrates and systematically investigated the effect of temperature on H_c, H_{eb}, and M_r/M_s of FeGa/IrMn bilayers. Linearly decreased H_c and H_{eb} with constant squareness are obtained with increasing measuring temperature for the samples grown on rigid Si substrates due to the thermal agitation. In contrast, for the sample with the PD parallel to the α_{31} direction, H_c decreases firstly and then increased at θ = 0° and 90° with increasing the temperature, because with increasing temperature the uniaxial anisotropy is reoriented from the α_{32} to α_{31} directions under a thermally induced compressive strain along the α_{32} direction. For the sample with the PD parallel to the α_{32} direction, H_c performs the similar tendency at θ = 0°, while H_c decreases with increasing the temperature at θ = 90°. The EB field of these two configurations decreases with increasing temperature. Meanwhile, the obvious changes in squareness of hysteresis loops were observed with changing temperature as a result of the strain induced magnetic anisotropy. A modified Stoner-Wohlfarth model was employed to simulate and explain the temperature modulation of the squareness in an EB system.

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