



## Article

# Large Perpendicular Exchange Energy in $Tb_xCo_{100-x}/Cu(t)/[Co/Pt]_2$ Heterostructures

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**Abstract:** In order to realize a perpendicular exchange bias for applications, a robust and tunable exchange bias is required for spintronic applications. Here, we show the perpendicular exchange energy (PEE) in the  $Tb_xCo_{100-x}/Cu/[Co/Pt]_2$  heterostructures. The structure consists of amorphous ferrimagnetic Tb–Co alloy films and ferromagnetic Co/Pt multilayers. The dependence of the PEE on the interlayer thickness of Cu and the composition of Tb–Co were analyzed. We demonstrate that the PEE can be controlled by changing the Cu interlayer thickness of  $0.2 < t_{Cu} < 0.3$  (nm). We found that PEE reaches a maximum value ( $\sigma_{Pw} = 1$  erg/cm<sup>2</sup>) at around  $x = 24\%$ . We, therefore, realize the mechanism of PEE in the  $Tb_xCo_{100-x}/Cu/[Co/Pt]_2$  heterostructures. We observe two competing mechanisms—one leading to an increase and the other to a decrease—which corresponds to the effect of Tb content on saturation magnetization and the coercivity of heterostructures. Sequentially, our findings show possibilities for both pinned layers in spintronics and memory device applications by producing large PEE and controlled PEE by Cu thickness, based on  $Tb_xCo_{100-x}/Cu/[Co/Pt]_2$  heterostructures.

**Keywords:** perpendicular magnetic anisotropy; ferrimagnet; perpendicular exchange bias; amorphous thin films; spintronic applications



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## 1. Introduction

The exchange bias (EB) phenomenon was discovered more than half a century ago by Meiklejohn and Bean [1]. EB can be observed through the exchange coupling between ferromagnet (FM)/antiferromagnetic (AFM) layers at the interface [2,3]. Utilizing a large perpendicular EB field as pinned layers in giant-magnetoresistive (GMR) devices, hard-disk drives (HDDs), magnetic random-access memory (MRAM) technologies, and magnetic tunnel junctions (MTJs) have been the subjects of intense attraction because of their potential in spintronic applications [4–9]. On the other hand, in memory device applications, controlling the perpendicular exchange energy (PEE) is a crucial factor [10–13].

The exchange anisotropy energy is generally revealed by the exchange energy,  $\sigma_{Pw}$ , which is the stabilizing energy per unit area of the FM/AFM or ferrimagnet (FIM)/FM interfaces,  $H_{ex} = \frac{J_k}{M_s t} = \frac{\sigma_{Pw}}{2M_s t}$ , where  $M_s$  and  $t_{FM}$  are the saturation magnetization and thickness of the FM layer, respectively [14,15]. However, typical AFM/FM systems indicate a limitation in attaining large EB fields (usually below 1 KOe) [16–18], which correlates to challenges in fabricating fine AFM crystals, controlling the AFM domain state, and uncompensated spin moments at the interface [16,17,19]. Thus, it seems that these cannot provide the reasonable necessities for future spintronic applications. Aside from FM/AFM systems, exchange bias also exists in ferrimagnet FIM/FIM [20] and the ferromagnet FIM/FM bilayer [21]. Amorphous rare earth-transition metal (RE-TM) multilayers exhibit strong perpendicular magnetic anisotropy (PMA) and robust coupling interactions at the interface [22,23].

In amorphous ferrimagnetic (FI) rare earth-transition metal (RE-TM) alloy films, there are two kinds of pair interactions, the antiparallel exchange between the RE-TM moments and the parallel exchange of the TM moments themselves; both interactions can provide a

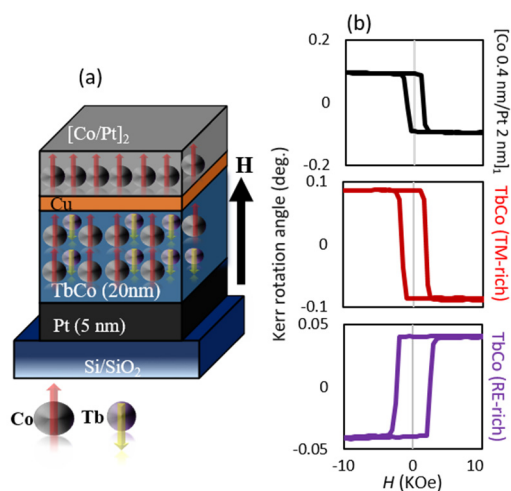
sufficiently strong interlayer coupling with the adjacent FM layer, and can hence provide a higher EB field—even for fully compensated interfaces [24,25].

Few kinds of research regarding the FIM/FIM, FM/FIM composite structures have been performed. Due to their potential for spintronic applications, more investigation is required to understand the mechanisms of these systems [20,26,27].

In this paper, we investigate the perpendicular exchange energy (PEE) between Tb–Co alloy films and Co/Pt multilayers by varying the Tb content of the Tb–Co layer. We observe that both EB and PEE can be tuned by introducing a Cu spacer layer. The PEE attains its maximum  $\sigma_{PW} = 1 \text{ erg/cm}^2$  at  $x = 24$ . We also describe the reason for high PEE at  $x = 24$  in the  $\text{Tb}_x\text{Co}_{100-x}(20)/\text{Cu}(t)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  heterostructures. Our findings prove that  $\text{Tb}_x\text{Co}_{100-x}(20)/\text{Cu}(t)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  heterostructures are efficient for spintronic applications, and they have advantages for manipulation by adjusting the Tb concentration.

## 2. Experimental Method

Figure 1a shows a schematic illustration of our  $\text{SiO}_2/\text{Pt}(5)/\text{Tb}_x\text{Co}_{100-x}(20)/\text{Cu}(t)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  (thicknesses in nm) ( $20 < x < 41$ ) samples, where the numbers in parentheses indicate the thickness in nanometers. The samples are deposited, using an ultrahigh-vacuum magnetron-sputtering system, onto a silicon substrate at room temperature. Co/Pt multilayers were sputtered, through dc sputtering, using two separate targets of platinum and cobalt at an argon-gas pressure level of 0.2 pascal.  $\text{Tb}_x\text{Co}_{100-x}$  films were prepared through co-sputtering using two separate targets of terbium and cobalt. The sample holder rotates during the deposition to ensure a uniform film composition. The thickness of the  $\text{Tb}_x\text{Co}_{100-x}$  layer is fixed at 20 nm. The composition of the films was measured using energy-dispersive X-ray analysis (EDX). The magnetic properties were measured at room temperature using the polar-magneto-optical Kerr effect (PMOKE) and a vibrating sample magnetometer (VSM).



**Figure 1.** (a) Schematic view of the sample layer structure; (b) out-of-plane magneto-optical Kerr effect.

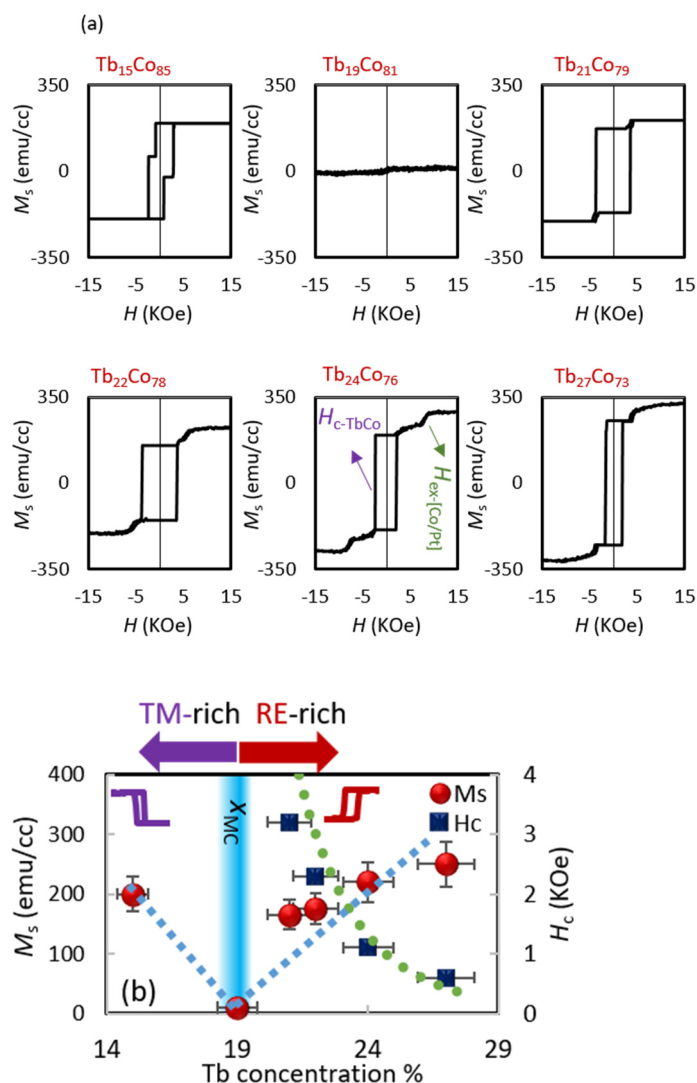
## 3. Results and Discussion

We first determined the magnetic properties of  $\text{Tb}_x\text{Co}_{100-x}(20)/\text{Cu}(0.2)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  systems. Figure 1b shows the hysteresis loops were measured using the polar-magneto-optical Kerr effect (PMOKE) at room temperature for three single-layer samples, a  $[\text{Co}/\text{Pt}]_2$  multilayer film, and Tb–Co films. All samples show the easy axis perpendicular to the film plane. The polarity of the Kerr rotation ( $\theta_K$ ) signals switches, which is consistent with a transition from being Co dominated to being Tb dominated in the magnetic moment.

(MOKE) signal for a single layer of  $[\text{Co}/\text{Pt}]$  and Tb–Co. The MOKE measurement wavelength is 690 nm for visible light. At 690 nm, only the magneto-optical Kerr effect of Co can be measured. This magneto-optical hysteresis of Co shows negative polarity, as shown in Co/Pt in Figure 1b. Similarly, in the TM-rich Tb–Co single-layer in

Figure 1b, since the Co in the Tb–Co layer is aligned in the magnetic field direction, the polarity of the hysteresis is negative, as in Co/Pt. On the other hand, in the RE-rich Tb–Co single-layer sample in Figure 1b, the polarity of hysteresis is negative because the Co is aligned in the opposite direction to the magnetic field.

Figure 2a shows the out-of-plane magnetic hysteresis loops were measured using the VSM at room temperature for  $Tb_xCo_{100-x}(20)/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  systems with different Tb concentrations. Here, we used the VSM to find the saturation magnetization of the sample, which is summarized in Figure 2b. Figure 2a shows a two-step switching loop, where the first and second loop switches at the low and high magnetic fields correspond to the Tb–Co and [Co/Pt] multilayers, respectively.



**Figure 2.** (a) The out-of-plane M–H loop for  $Tb_xCo_{100-x}/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  heterostructures, and (b)  $M_s$  and  $H_c$  as a function of Tb concentration.

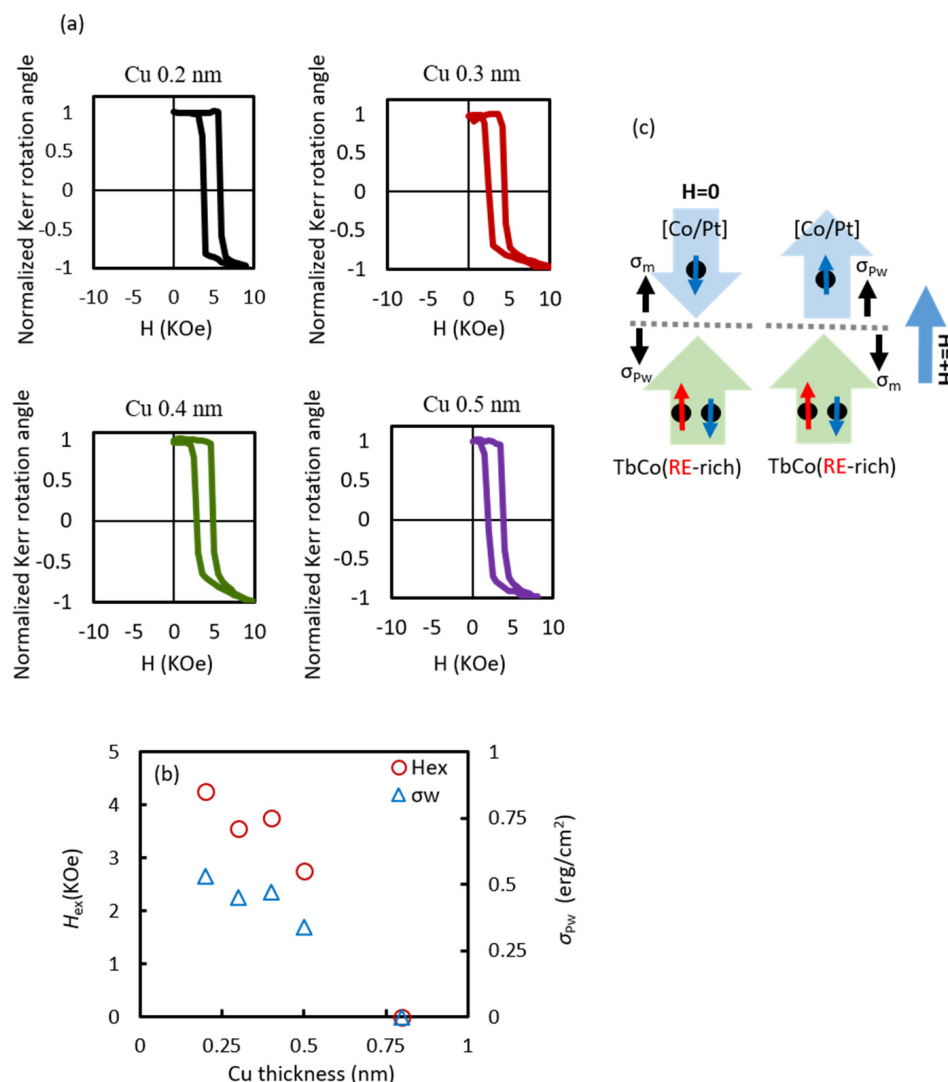
Figure 2b shows the coercive fields ( $H_c$ ) and the saturation magnetizations ( $M_s$ ) of  $Tb_xCo_{100-x}(20)/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  films at different compositions. From the magnetization curve of the  $SiO_2/Pt(5)/Tb_xCo_{100-x}(20)/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  heterostructures, it is seen that the saturation magnetization the  $M_s$  of  $Tb_xCo_{100-x}$  reaches its magnetization compensation composition point at  $x_C \sim 19$ . While the  $M_s$  is at its minimum, the coercive fields reach their maximum at the magnetic compensation composition [28–31]. On

the other hand, the perpendicular magnetic anisotropy ( $K_u$ ) can be calculated using the following equation:

$$K_u \approx \alpha M_s H_c \quad (1)$$

where the  $\alpha$  is constant for all samples, since all heterostructures are prepared under the same conditions. By increasing the Tb concentration the  $K_u$  value decreases for all samples, which is in good agreement with the previous report [26,32,33].

To realize the insertion layer effect on  $H_{ex}$  and the  $\sigma_{Pw}$ , Figure 3a shows the out-of-plane minor loops (+15 KOe  $\Rightarrow$  0 Oe  $\Rightarrow$  +15 KOe) for the  $Tb_{21}Co_{79}/Cu(t_{Cu})/[Co(0.4)/Pt(2)]_2$  heterostructures ( $0.2 < t_{Cu} < 1$  nm). This hysteresis loop is shifted away from the zero-field axis to  $H = +H_{ex}$ , and the width of the loop is  $2 H_c$ , where  $H_c$  is the coercive field of a  $[Co/Pt]_2$  layer. The hysteresis curve on the high magnetic field side, shown in Figure 2, shows the magnetization reversal of the Co/Pt layer, as evidenced by the negative polarity of the minor loop, as shown in Figure 3a. If this was the result of RE-rich Tb–Co, the polarity would be positive.



**Figure 3.** (a) Minor loops of the  $Tb_{21}Co_{79}/Cu(t_{Cu})/[Co(0.4)/Pt(2)]_2$  heterostructures with various insertion layer thickness; (b) changes in the unidirectional anisotropy constant,  $\sigma_{Pw}$ , and exchange anisotropy,  $H_{ex}$ , as a function of Cu thickness; (c) schematic illustration of the magnetic configuration at the interface of the  $Tb_xCo_{100-x}/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  heterostructures.

The Cu interlayer, with a thickness of  $t_{\text{Cu}} = 0.2\text{--}1$  nm, was employed to tune the perpendicular exchange energy (PEE),  $\sigma_{\text{Pw}}$ . The optimization of the insertion layer thickness is necessary to control the PEE value because the removal of the Cu layer causes the exchange coupling to become very large; therefore, it is impractical for the observation of the shift of the hysteresis loop. Ideally, the effect of the Cu insertion causes the intermixing of Tb–Co–Co/Pt that appears during the sputtering process to decrease. Hence, inserting thin Cu layers allows for the improvement of the interface, thus enhancing the effective anisotropy of the stack [34]. Figure 3b shows that by increasing the  $t_{\text{Cu}}$  over 0.5 nm, the  $H_{\text{ex}}$  and PEE  $\sigma_{\text{Pw}}$  monotonically decrease, which is in agreement with previous reports [35,36]; thus, the optimum  $H_{\text{ex}}$  and  $\sigma_{\text{Pw}}$  values were 4.25 kOe and 0.54 erg/cm<sup>2</sup> at  $t_{\text{Cu}} = 0.2$  nm, respectively. As a result, it is clearly demonstrated that the PEE can be controlled by Cu thickness, which indicates additional suitability for memory device applications [10,11].

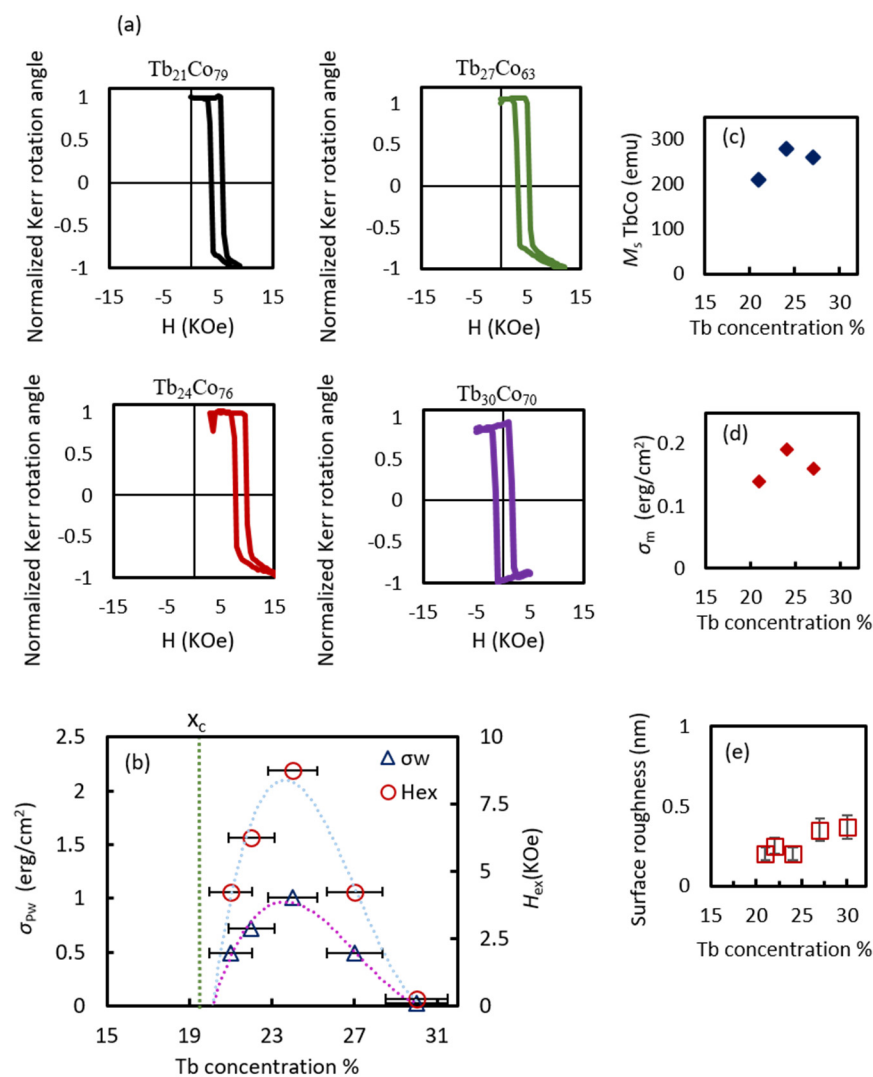
Figure 3c shows a schematic illustration of the magnetic configuration at the interface of the  $\text{Tb}_x\text{Co}_{100-x}/\text{Cu}(0.2)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  heterostructures. As shown by the zero magnetic field in the model diagram, the magnetization of the Co/Pt layer and the net magnetization of the Tb–Co layer are opposite from one another; therefore, the static magnetic energy at the interface increases. However, since the Co in the Co/Pt layer and the Co in the Tb–Co layer are both oriented in the same direction, the interfacial domain wall energy is low. On the other hand, in the high magnetic field in the model diagram, the static magnetic energy at the interface decreases because the Co/Pt layer is inverted, and conversely, the interfacial domain wall energy increases. In general, when the Co/Pt layer and the Tb–Co layer are directly heterojunctioned, the interfacial domain wall energy is overwhelmingly larger than the interfacial static magnetic energy; thus, it can only be used only for a large exchange bias application. However, it was found that by inserting a small amount of Cu at these interfaces, the interfacial domain wall energy can be controlled and reduced to the desired value. As a result, it can be applied to memory applications that also utilize interfacial static magnetic energy.

To define the EB for each of the heterostructures, the out-of-plane minor loops were measured as shown in Figure 4a. Figure 4b summarized  $\sigma_{\text{Pw}}$  and the magnitude of the  $H_{\text{ex}}$  field, respectively, as a function of the Tb content in the  $\text{Tb}_x\text{Co}_{100-x}/\text{Cu}(0.2)/[\text{Co}(0.4)/\text{Pt}(2)]_2$  heterostructures at room temperature. Between 23 and 25 at.% Tb, the PEE seems to reach its maximal value; toward lower and higher amounts of Tb, some reduction appears. In the first region, both the  $H_{\text{ex}}$  and  $\sigma_{\text{Pw}}$  values increase when the Tb is among  $19 < x < 24$  atomic percent. Contrarily, in the second region—by increasing the Tb content from  $x = 24$  [31]—both the  $H_{\text{ex}}$  and  $\sigma_{\text{Pw}}$  values decrease and become zero at around  $x = 30$ .

In previous studies, it was reported that the EB field reaches its maximum at the compensation point since the compensated sublattices of the FIM film hold no frustrated bonds at the interface to the FM layer [25,37]. However, this behavior cannot be explained directly in our system.

Firstly, this behavior can be explained by taking into account the variation in the exchange energies of the Co–Co pair with changing RE content [38]. Accordingly, the exchange coupling between the Co–Co pairs is the strongest when compared to other pairings ( $J_{\text{Co-Co}} > J_{\text{Co-Tb}} > J_{\text{Co-Pt}}$ ). The  $J_{\text{Co-Co}}$  is present at the interface between Tb–Co and [Co/Pt]; therefore, a maximum PEE appears at a lower Tb concentration. Increasing the Tb concentration can decrease the number of Co atoms at the interface, and occasionally it can cause the reduction in the exchange coupling between Co–Co pairs at the interface [21,39].

Moreover, by increasing the Tb content,  $H_c$  decreases while  $M_s$  increases, which is shown in Figure 2b. Therefore, the variation in the  $\sigma_{\text{Pw}}$  value in the reduction in  $H_c$  is likely connected with a smaller perpendicular exchange energy,  $\sigma_{\text{Pw}}$ , in the ferrimagnet, which should result in smaller perpendicular exchange energy.



**Figure 4.** (a) Minor loops of the  $Tb_xCo_{100-x}/Cu(0.2)/[Co(0.4)/Pt(2)]_2$  heterostructures; (b) changes in perpendicular exchange energy,  $\sigma_{Pw}$ , and exchange anisotropy,  $H_{ex}$ , as a function of Tb composition; (c) Tb–Co magnetization as a function of the Tb content; (d) change in magnetostatic energy as a function of Tb composition; (e) surface roughness plotted as a function of Tb concentration.

To clarify the variation in  $\sigma_{Pw}$ , the exchange energy can be calculated by the relation in Equation (2). Hence, the total magnetic energy at the interface can be explained by the following equation:

$$E = \sigma_{Pw} + \sigma_m \quad (2)$$

Here,  $\sigma_{Pw} = \sigma_{iw} + \sigma_A$ , where the first term is the interfacial domain wall energy generated between the Co/Pt layer and the Tb–Co layer; the second term,  $\sigma_m$ , is the static magnetic energy generated between the Co/Pt layer and the Tb–Co layer; and the third term,  $\sigma_A$ , is the magnetic anisotropy generated between the Co/Pt layer and the Tb–Co layer. The interfacial domain wall energy obtained from the inverting magnetic field  $H_{EX}$  of the Co/Pt layer is  $\sigma_{Pw}$ .

The anisotropic energy is very small because the Cu intermediate layer greatly attenuates the exchange force between the Co/Pt layer and the Tb–Co layer. Therefore, this  $\sigma_A$  term can be negligible [40]. To realize the mechanism of the PEE at the interface, we extracted the magnetization information of the samples in Table 1 and summarized them in Figure 4c.

**Table 1.** Summarized magnetic properties of Tb–Co/[Co/Pt]<sub>2</sub> multilayers.

FIM Composition	Ms-Tb-Co (emu/cm <sup>3</sup> )	Ms-[Co/Pt] (emu/cm <sup>3</sup> )
Tb <sub>21</sub> Co <sub>79</sub>	210	1580
Tb <sub>24</sub> Co <sub>76</sub>	280	1430
Tb <sub>27</sub> Co <sub>73</sub>	260	1550

Figure 4d shows the magnetostatic energy of the Tb<sub>x</sub>Co<sub>100-x</sub>. It is seen that the magnetostatic energy shows the same curvature, which is in good agreement with Figure 4c. Furthermore, the value of magnetostatic energy is considerable and plays an important role in this system [41].

Since EB is an interfacial phenomenon, the surface roughness may affect the magnitude of the exchange bias [19,42,43]. Hence, the effect of the Tb content on the surface morphology of Tb<sub>x</sub>Co<sub>100-x</sub>/Cu(0.2)/[Co(0.4)/Pt(2)]<sub>2</sub> heterostructures was investigated using AFM. Figure 4e shows a flat surface for Tb<sub>24</sub>Co<sub>76</sub> in comparison to the other composition. Surface roughness (*R<sub>a</sub>*) for Tb–Co increases with increasing Tb concentration. Therefore, the variation in the exchange bias field as a function of the Tb composition might be related to the interface roughness induced by changes in growth conditions, depending on the Tb content of the alloy.

For the Tb<sub>x</sub>Co<sub>100-x</sub> samples, both *H<sub>ex</sub>* and *σ<sub>PW</sub>* reach a maximum value of *H<sub>ex</sub>* = 8.75 kOe and *σ<sub>PW</sub>* = 1 (erg/cm<sup>2</sup>) at *x* = 24, which are significantly larger than what was observed in the ordinary AFM/FM and FM/FM systems [17,27,36,44,45].

#### 4. Summary

In summary, we have systematically investigated the perpendicular exchange bias and perpendicular exchange energy (PEE) *σ<sub>PW</sub>* of Tb<sub>x</sub>Co<sub>100-x</sub>/Cu(*t<sub>Cu</sub>*)/[Co(0.4)/Pt(2)]<sub>2</sub> (20 < *x* < 30) heterostructures. We replaced the commonly used AFM pinned layer with the ferrimagnet pinning layer. The interlayer thickness and FIM composition of the Tb–Co layer were optimized to obtain large *H<sub>ex</sub>* and *σ<sub>PW</sub>*. The advantage of using amorphous RE-TM alloys as a pinned layer is the tunable magnetic properties that depend strongly on composition. The PEE reached a maximum *σ<sub>PW</sub>* = 1 (erg/cm<sup>2</sup>) around *x* = 24 at.%, at room temperature. In this system, we observed two competing mechanisms—one leading to an increase and the other to a decrease—which corresponds to the effect of Tb content on saturation magnetization and the coercivity of heterostructures. The developed FIM/FM films, with a perpendicular exchange bias and a large PEE, will be greatly beneficial in spintronic applications, such as magneto-optical memory and high areal density recording technology.

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