

## Supplementary Information

### Field-Free switching of perpendicular magnetization through spin-orbit torque in FePt/[TiN/NiFe]<sub>5</sub> multilayers

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#### I. The AHE, crystallographic structures and current induced switching loops of FePt/[TiN/NiFe]<sub>3</sub> multilayers

Similar to the stacks of FePt/[TiN/NiFe]<sub>5</sub> multilayers, XRD image (Fig.S1 b) shows only weak FePt (001) peaks for FePt/[TiN/NiFe]<sub>3</sub> RT, indicating poor (001) texture and chemical ordering. For FePt/[TiN/NiFe]<sub>3</sub> HT, the chemical ordering increases as well as chemical ordering. Moreover, epitaxial growth of TiN/NiFe on the FePt film is also observed for FePt/[TiN/NiFe]<sub>3</sub> HT. As for AHE results (Fig.S1 a), it also shows similarity for FePt/[TiN/NiFe]<sub>3</sub> as compared to FePt/[TiN/NiFe]<sub>5</sub> such as the weaker PMA of FePt/[TiN/NiFe]<sub>3</sub> RT. Moreover, Figure S1 c and d give the current induced switching loops of FePt/[TiN/NiFe]<sub>3</sub>. All the results are almost consistent with FePt/[TiN/NiFe]<sub>5</sub> including the change of rotation direction of switching loops under different  $H_x$ . The main difference is switching polarity under  $H_x=0$  Oe for FePt/[TiN/NiFe]<sub>3</sub> HT. This indicates different inherent  $H_{in}$ , which may be affected by the layer numbers of TiN/NiFe.

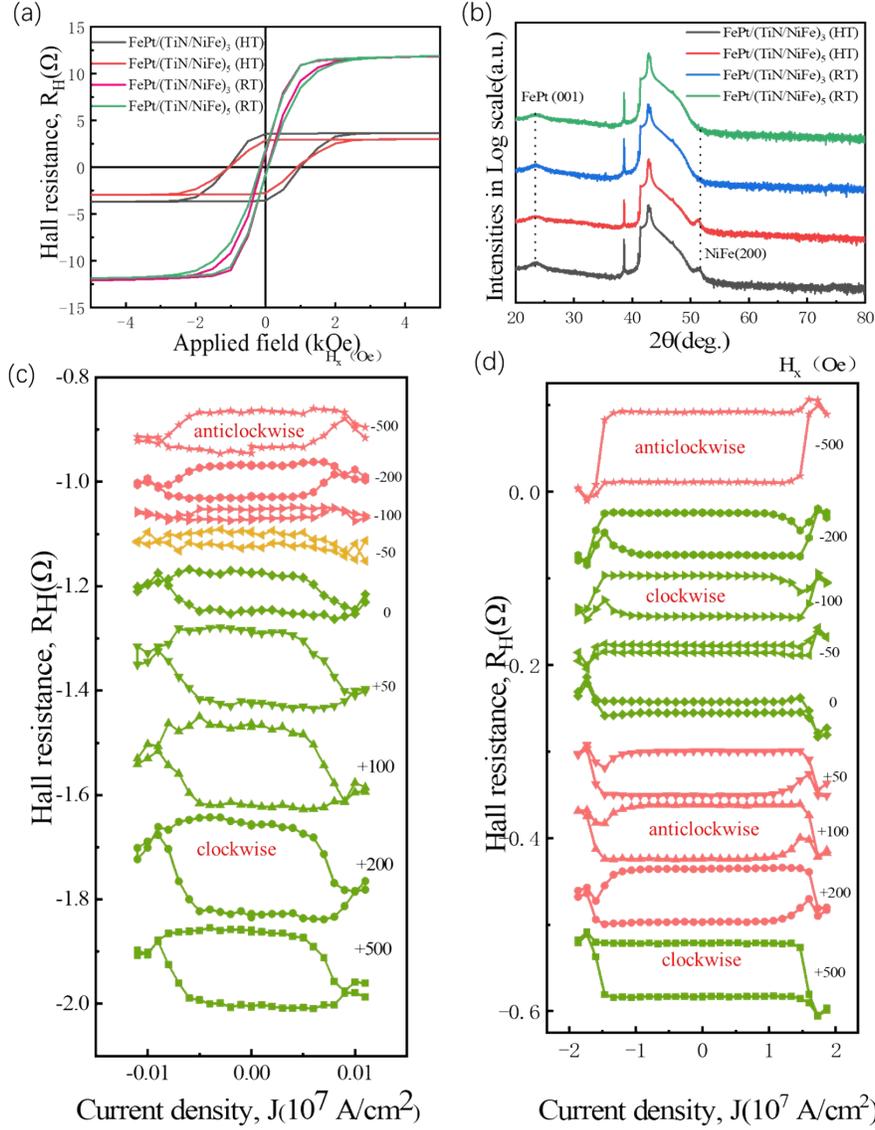


Figure S1 (a) AHE loops of FePt/[TiN/NiFe]<sub>n</sub> multilayers, (b) the corresponding XRD image of the FePt/[TiN/NiFe]<sub>3</sub> multilayers, (c) current-induced magnetization switching of FePt films with different external fields  $H_x$  of FePt/[TiN/NiFe]<sub>3</sub> RT (d) current-induced magnetization switching of FePt films with different external fields  $H_x$  of FePt/[TiN/NiFe]<sub>3</sub> HT

## II. Investigations on IEC in FePt/[TiN/NiFe]<sub>n</sub>

To scrupulously investigate the IEC in FePt/[TiN/NiFe]<sub>n</sub>, we have studied the performance of FePt/TiN/NiFe trilayers, in which different thicknesses of TiN(0, 1, 2, 3nm) are used. The results are shown in Fig. S2. For devices with TiN 0 nm, no deterministic switching is observed at  $H_x=0$  Oe and the switching behavior under different  $H_x$  is similar to the behavior of single FePt, indicating the

inexistence of IEC (Fig. S2 c). While for the devices with TiN 1nm (Fig. S2 d), the field-free switching is realized and by varying external field (from 100 Oe to -100 Oe) the transition of switching polarity between clockwise and anticlockwise only happens for once. Such behavior is consistent with the ferromagnetic coupling in our model. As for the devices with TiN 2, 3nm (Fig. S2 e, f), the switching polarities change for three times with the  $H_x$  from 500 Oe(200 Oe) to -500 Oe (-200 Oe), which is similar to the behavior of FePt/[TiN/NiFe]<sub>5</sub> HT (sample 2 in main manuscript). This characteristic is in agreement with the antiferromagnetic coupling. Those experiments prove that with varying the thickness of space layer the oscillation between ferromagnetic and antiferromagnetic coupling happens, which is a typical attribute of IEC.

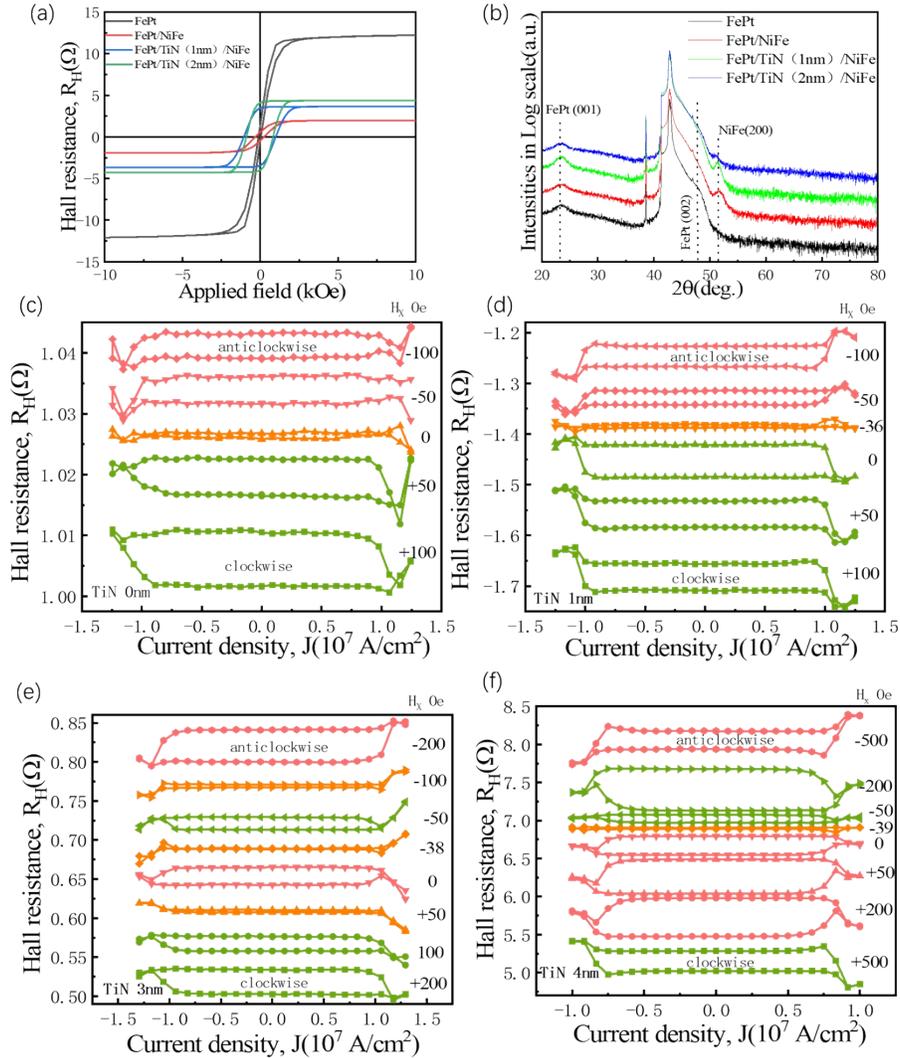


Figure S2 (a) the AHE loops for different structures, (b) the corresponding XRD results, the current induced magnetization switching for FePt under varying in-plane field  $H_x$  of samples with different TiN thickness (c) TiN 0nm, (d) TiN 1nm, (e) TiN 2nm, (f) TiN 3nm, respectively

Furthermore, we will discuss the IEC in FePt/[TiN/NiFe]<sub>n</sub>. According to Bruno's study [1], the oscillations of IEC depend on both the thickness of space layer and ferromagnetic layer. As a result, the IEC between FePt and [TiN/NiFe]<sub>n</sub> is affected by the thickness of TiN and NiFe, the number of n and the IEC of NiFe/TiN/NiFe, which is a complex question. In order to simplify the effect of IEC on FePt SOT switching, in this paper, we mainly focus on the effect of the thickness of TiN and the number of n, and neglect the other factors.

Following this, Table 1 summaries the characteristics of IEC for different structures. In details, the thickness of TiN in main manuscript is that 1nm for sample 3 (FePt/[TiN/NiFe]<sub>5</sub> RT) and 2 nm for sample 2 (FePt/[TiN/NiFe]<sub>5</sub> HT) due to formation temperature. The results show that the thickness of TiN and n in [TiN/NiFe]<sub>n</sub> can affect the type of IEC and thus influence the critical values for transition of switching polarities.

Table 1 the comparison of IEC effects for different structures

<i>Structure</i>	<i>Type of IEC</i>	<i>Change times of switching polarities</i>	<i>Critical values for transition of switching polarities (Oe)</i>
FePt	~	1	0
FePt/NiFe	~	1	0
FePt/TiN(1nm)/NiFe	FM	1	-36
FePt/TiN(2nm)/NiFe	AFM	2	1 <sup>st</sup> 50, 2 <sup>st</sup> -38, 3 <sup>st</sup> -100
FePt/TiN(3nm)/NiFe	AFM	3	1 <sup>st</sup> 200~500, 2 <sup>st</sup> -39, 3 <sup>st</sup> -200~-500
FePt/[TiN(1nm)/NiFe] <sub>3</sub> HT	AFM	3	1 <sup>st</sup> 200~500, 2 <sup>st</sup> 0~50, 3 <sup>st</sup> -200~-500
FePt/[TiN(2nm)/NiFe] <sub>3</sub> RT	FM	1	-50
FePt/[TiN(1nm)/NiFe] <sub>5</sub> HT	AFM	3	1 <sup>st</sup> 200, 2 <sup>st</sup> 0~50, 3 <sup>st</sup> -200~-500
FePt/[TiN(2nm)/NiFe] <sub>5</sub> RT	FM	1	-50

### III. The possible source of $H_{in}$

We firstly check the FePt/TiN interface. As shown in Fig S3 a, there is no magnetization switching loops at  $H_x=0$  Oe, indicating the FePt/TiN interface are not responsible for such inherent field. Then, the magnetic properties of FePt film are studied. Two FePt samples with different deposition temperature (300°C or 500°C) are prepared and Fig. S3 d shows their AHE loops, which indicating the coercivity of 500°C deposited FePt is much larger than that of 300°C deposited sample. Following this, the two FePt/TiN/NiFe samples are fabricated, during this process the sputtering conditions for TiN and

NiFe layer keep the same. AHE results (Fig. S3 e) show that the sample with 500°C FePt still has larger coercivity, while coercivity of the sample with 300°C FePt is close to the value of FePt/[TiN/NiFe]<sub>5</sub> HT (sample 2 in main manuscript). Fig. S3 b shows the switching loops for the sample with 500°C FePt/TiN(2nm)/NiFe. By changing the direction of NiFe film, the switching polarity of FePt film is changeable at  $H_x=0$  Oe. Such characteristics is a typical anti-ferromagnetic coupling and there is no inherent field. On the contrary, the inherent field appears in sample with 300°C FePt (Fig. S3 c). Thus, we infer the source of inherent field is associated with the magnetic properties of FePt film and may originate from the complex coupling in FePt/TiN/NiFe system, which needs further investigation.

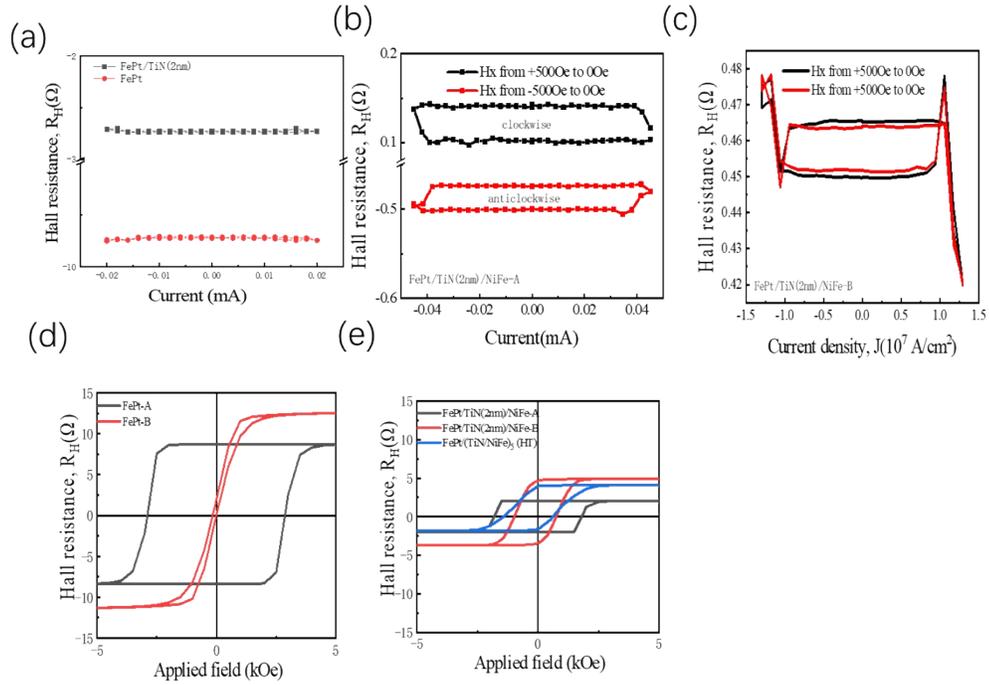


Figure S3 current-induced magnetization switching of FePt films at  $H_x=0$  Oe of (a) FePt and FePt/TiN bilayers; the switching loops under  $H_x=0$  Oe after using an in-plane 500 Oe or -500 Oe fields to saturate NiFe for (b) FePt/TiN(2nm)/NiFe-A, in which FePt is deposited at 500°C, (c) FePt/TiN(2nm)/NiFe-B, in which FePt is deposited at 300°C; (d) AHE loops for FePt with different deposition temperature, FePt-A 500°C, FePt-B 300°C; (e) AHE loops for different samples

#### IV. The switching models for sample 2 and sample 3

The change of  $H_E$  relates to the remanent magnetization of NiFe layer. With varying the external field  $H_x$ , the changeable remanent magnetization of NiFe may result in the variable  $H_E$ , which exists both in sample 2 and 3. However, the coupling type is different, that is antiferromagnetic coupling for sample 2 and ferromagnetic coupling for sample 3. For antiferromagnetic coupling, the effective field

$H_E$  performs opposite direction compared to the magnetic direction of NiFe layer, while the direction of  $H_E$  is the same as the direction of NiFe for ferromagnetic coupling. As a result, the different directions of  $H_E$  combined with  $H_{in}$  and external field  $H_x$  cause the different switching characteristics for sample 2 and 3.

In details, with varying  $H_x$  from -500 Oe to 500 Oe the switching processes for sample 2 are shown in Fig. S4. When  $H_x$  is smaller than -50 Oe, the large remanent magnetization of NiFe induces the maximum  $H_{E_{max}}$  (corresponding to A to C sites). In those sites, the  $H_x$  and  $H_{in}$  have the same direction, which is opposite to  $H_{E_{max}}$ . Decreasing  $|H_x|$ , the relationship between  $|H_x| + |H_{in}|$  and  $|H_{E_{max}}|$  varies and switching direction changes for once. At site D ( $H_x$  between -50 Oe to 0 Oe), the decreasing remanent magnetization of NiFe causes the decreased  $H_{E_{min1}}$  and the  $|H_x| + |H_{in}| = |H_{E_{min1}}|$  determines the disappearance of switching loop. When  $H_x$  is equal to 0 Oe (site E), the further decreased remanent magnetization results in smaller  $H_{E_{min0}}$  and the field-free switching is obtained due to  $|H_{in}| > |H_{E_{min0}}|$ . Between D and E site, the second change for switching direction happens. On the contrary, when varying  $H_x$  from 50 Oe to 500 Oe, the magnetization of NiFe changes the direction. The  $H_{-E}$  and  $H_{in}$  have the same direction, opposite to  $H_x$ . Similarly, the relationship between  $|H_{-E_{max}}| + |H_{in}|$  and  $|H_x|$  varies due to changeable  $|H_x|$  and the third change for switching direction is observed.

As for sample 3, the switching processes are shown in Fig. S5, under  $H_x$  from -500 Oe to 500 Oe. When  $H_x$  is smaller than -100 Oe,  $H_x$  and  $H_E$  have the same direction, opposite to  $H_{in}$ . The sum of  $|H_x| + |H_{E_{max}}|$  is always larger than  $|H_{in}|$  and the switching direction keeps unchangeable. Decreasing  $H_x$  to -50 Oe (site B), the smaller remanent magnetization of NiFe corresponds to smaller  $H_{E_{min1}}$  and  $|H_x| + |H_{E_{min1}}| = |H_{in}|$  determines the disappearance of switching loop. When  $H_x$  is equal to 0 Oe (site C), the field-free switching is obtained due to  $|H_{in}| > |H_{E_{min0}}|$ . For  $H_x$  varying from 50 Oe to 500 Oe, the magnetic direction of NiFe changes and  $H_E$ ,  $H_x$ ,  $H_{in}$  have the same direction. In this case, no matter how  $H_E$  changes with respect to remanent magnetization of NiFe the switching polarities always keep the same. As a result, the change for switching directions can be only observed for once.

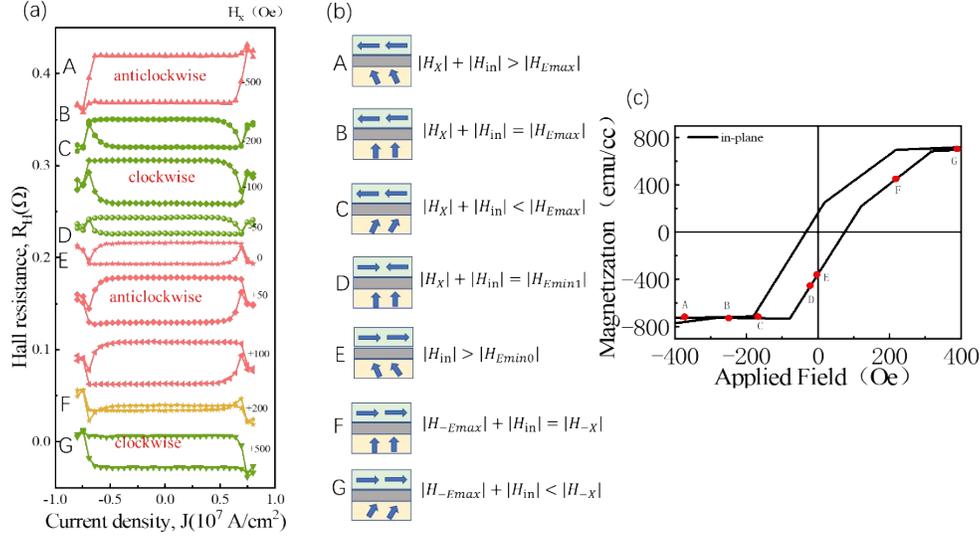


Figure S4 (a) current-induced magnetization switching of FePt films with different external fields  $H_x$  of sample 2, (b) the corresponding model of switching process with different  $H_x$ , (c) the remanent magnetization states of NiFe with different  $H_x$

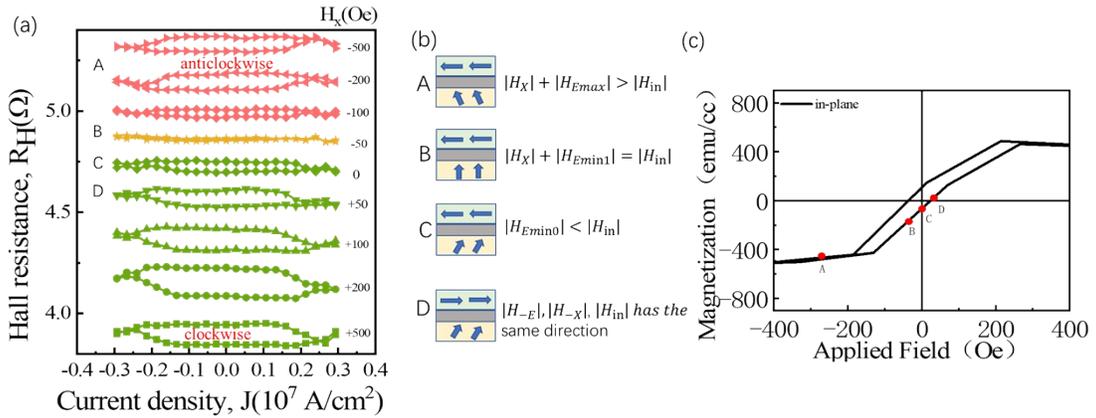


Figure S5 (a) current-induced magnetization switching of FePt films with different external fields  $H_x$  of sample 3, (b) the corresponding model of switching process with different  $H_x$ , (c) the remanent magnetization states of NiFe with different  $H_x$

## V. The memristive behavior of sample 2 with increasing counts of positive pulses from 20 to 80

A series of current pulses are applied with varying counts of positive pulses from 20 to 80, and similarly  $R_H$  varies gradually with the increasing pulses numbers, as shown in Fig. S6. Such a stably memristive characteristics of our devices shows good application as a synapse in a self-adaptive network.

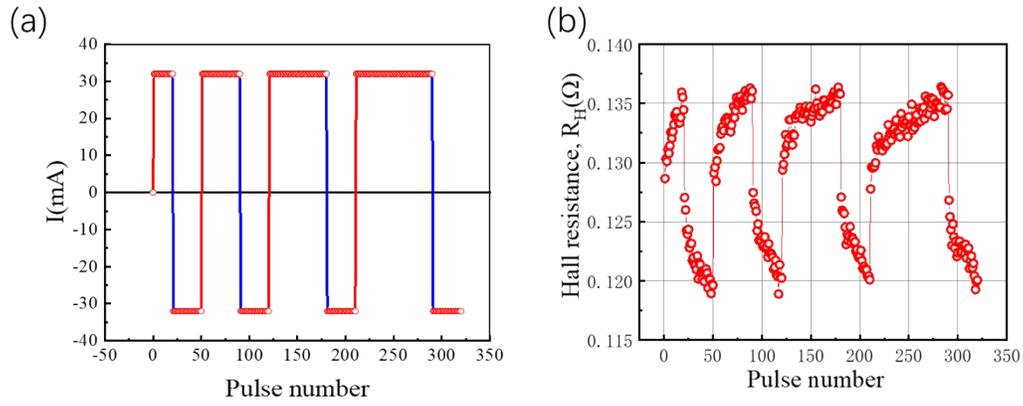


Figure S6 (a) the applied train of current pulses with  $H_x=200$  Oe, (b) the corresponding response of  $R_H$

## REFERENCES

- [1] P. Bruno, Oscillations of interlayer exchange coupling vs. ferromagnetic-layers thickness, *EPL*, **1993**, 23: 615.