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Solar manipulations of perpendicular magnetic anisotropy for flexible spintronics

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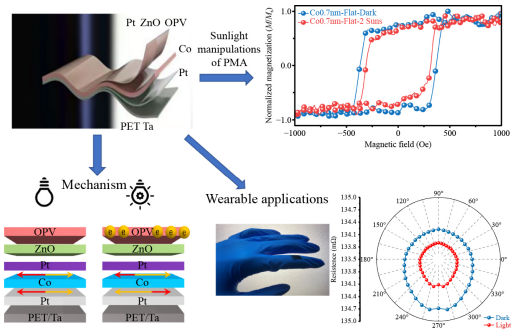
ABSTRACT

Flexible electronics/spintronics attracts researchers' attention for their application potential abroad in wearable devices, healthcare, and other areas. Those devices' performance (speed, energy consumption) is highly dependent on manipulating information bits (spin-orientation in flexible spintronics). In this work, we established an organic photovoltaic (OPV)/ZnO/Pt/Co/Pt heterostructure on flexible PET substrates with perpendicular magnetic anisotropy (PMA). Under sunlight illumination, the photoelectrons generated from the OPV layer transfer into the PMA heterostructure, then they reduce the PMA strength by enhancing the interfacial Rashba field accordingly. The coercive field (H_c) reduces from 800 Oe to 500 Oe at its maximum, and the magnetization can be switched up and down reversibly. The stability of sunlight control of magnetization reversal under various bending conditions is also tested for flexible spintronic applications. Lastly, the voltage output of sunlight-driven PMA is achieved in our prototype device, exhibiting an excellent angular dependence and opening a door towards solar-driven flexible spintronics with much lower energy consumption.



Keywords interfacial magnetoelectric coupling, perpendicular magnetic anisotropy, deterministic magnetization reversal, photovoltaic control of magnetism

1 Introduction

As a rapidly growing technology, flexible electronics attracts researchers' attention for its application potential abroad in wearable devices, healthcare, and other areas requiring flexible operating conditions [1–10]. In particular, flexible spintronics is fast, compact, and energy-saving compared with traditional flexible electronics, where the information is generated, transmitted, and manipulated by changing the spin orientation, instead of moving the charge carrier. Nevertheless, controlling the spin orientation on a curvature surface is challenging because a magnetic head with the localized magnetic field (H-field) cannot be applied in this sequence. Meanwhile, the complex strain/stress also influences the spin alignment, resulting in an unstable performance of devices [11, 12]. The spin-polarized current is recently developed to tune the spin orientation in various ferro-

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magnetic heterostructures, including ones on flexible substrates [13–16]. However, driving spin through current leads to inevitable charge movement, which eventually causes large energy consumption and loss of the advantage of spintronics. Considering the relatively small charge-to-spin ratio, the energy consumption of current-driven flexible spintronics is greater than expected.

In past decades, voltage control of magnetic anisotropy (VCMA) has been developed to overcome the weakness of current-driven spintronic devices, where the energy consumption can be significantly reduced by order of magnitudes by decreasing the current [17–27]. For example, researchers found out that the interfacial charge accumulation will shift the Fermi level of ferromagnetic thin films/heterostructures like exchange bias, perpendicular magnetic anisotropy (PMA) [28–31], synthetic antiferromagnetic (SAF) [18, 32–34], etc., and change its ferromagnetism such as saturation magnetization, magnetic anisotropy, and interlayer coupling, accordingly. In our previous research, we realized an ionic gating control of PMA and SAF by reversing the magnetization 90 or 180 degrees at a small voltage (< 4 V) on both rigid and flexible substrates, showing the potential of VCMA on flexible spintronic devices [28, 32]. Nonetheless, the ions' slow movement limits the tuning rate, and the interfacial chemical reaction is inescapable due to the considerable ionic accumulation. Recently, we exploited a sunlight control of interfacial ferromagnetism in photovoltaic/magnetic heterostructures with a fast response and clean interface without chemical corrosion [35–39]. For instance, a sunlight control of near 180-degree deterministic magnetization reversal is realized in photovoltaic/PMA heterostructure with the assistance of the magnetic bias [40].

This work establishes an organic photovoltaic (OPV)/ZnO/Pt/Co/Pt heterostructure on flexible substrates (PET, Mica) with PMA. The PMA structure is magnetron-sputtered onto flexible substrates with OPVs and ZnO layers spin-coated onto it, as shown in Figs. 1(a) and (b). The ZnO layer increases the photoelectron transmitting rate and improves the photon-to-electron conversion efficiency (PCE). Under sunlight illumination, the photoelectrons generated from the OPV layer transfer into the PMA heterostructure, then they reduce the PMA strength by enhancing the interfacial Rashba field accordingly. Thereby, the coercive field (H_c) reduces from 800 Oe to 500 Oe at its maximum, which means that the magnetization can be switched up and down reversibly under an appropriate magnetic field. The stability of sunlight control of magnetization reversal under various bending conditions is also tested for flexible spintronic applications. Lastly, the voltage output of sunlight-driven PMA is achieved in our prototype device, exhibiting an excellent angular dependence and opening a door towards solar-driven flexible spintronics with much lower energy consumption.

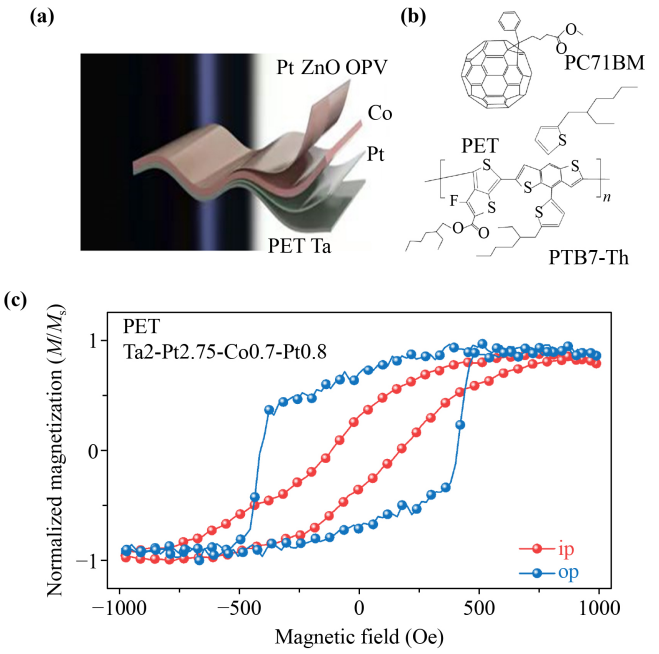

Fig. 1 The schematic of sunlight photovoltaic gating control of perpendicular magnetic anisotropy heterostructure on flexible substrates and in situ VSM measurement of the PMA heterostructures on Mica and PET substrates. **(a)** The schematic of organic photovoltaic/ZnO/Pt/Co/Pt/Ta/PET flexible heterostructure. **(b)** The molecular nano-structure of the acceptor (PC71BM) and donor (PTB7-Th). **(c)** The in-plane (red) and out-of-plane (blue) magnetic hysteresis loops on the PET substrate correspondingly.

2 Experimental section

2.1 Device fabrication

Pt (0.8 nm)/Co (x nm)/Pt (2.75 nm) multilayer heterostructures were deposited onto Mica and PET flexible substrates by a DC magnetron sputtering under high vacuum (1×10^{-7} Torr) at room temperature. The layer thickness was controlled with an integrated quartz crystal microbalance in a sputtering system during the deposition process. The 20 μm thick mica and 50 μm thick PET substrates were bought from Taiyuan Co. Jilin, China. Then, ZnO layers were spin-coated onto PMA heterostructure with 5000 rmp rotation speed for 30 s. The organic photovoltaic donor materials were PTB7-Th (poly[4,8-bis(5-(2-ethylhexyl) thiophene-2-yl) benzo[1,2-b:4,5-b']dithiophene-co-3-fluorothieno[3,4-b] thiophene-2-carboxylate])). The acceptor material was PC71BM ([6,6]-phenyl C71 butyric acid methyl ester). PTB7-Th and PC71BM were purchased from 1-Material Chemscitech Inc. (Canada). The solution was stirred overnight at 75 °C before the active organic layer was fabricated. Active layers were spin-coated by the polymer solution on the substrate in an ambient atmosphere at 2000 rpm for 30 s. PTB7-Th and PC71BM were

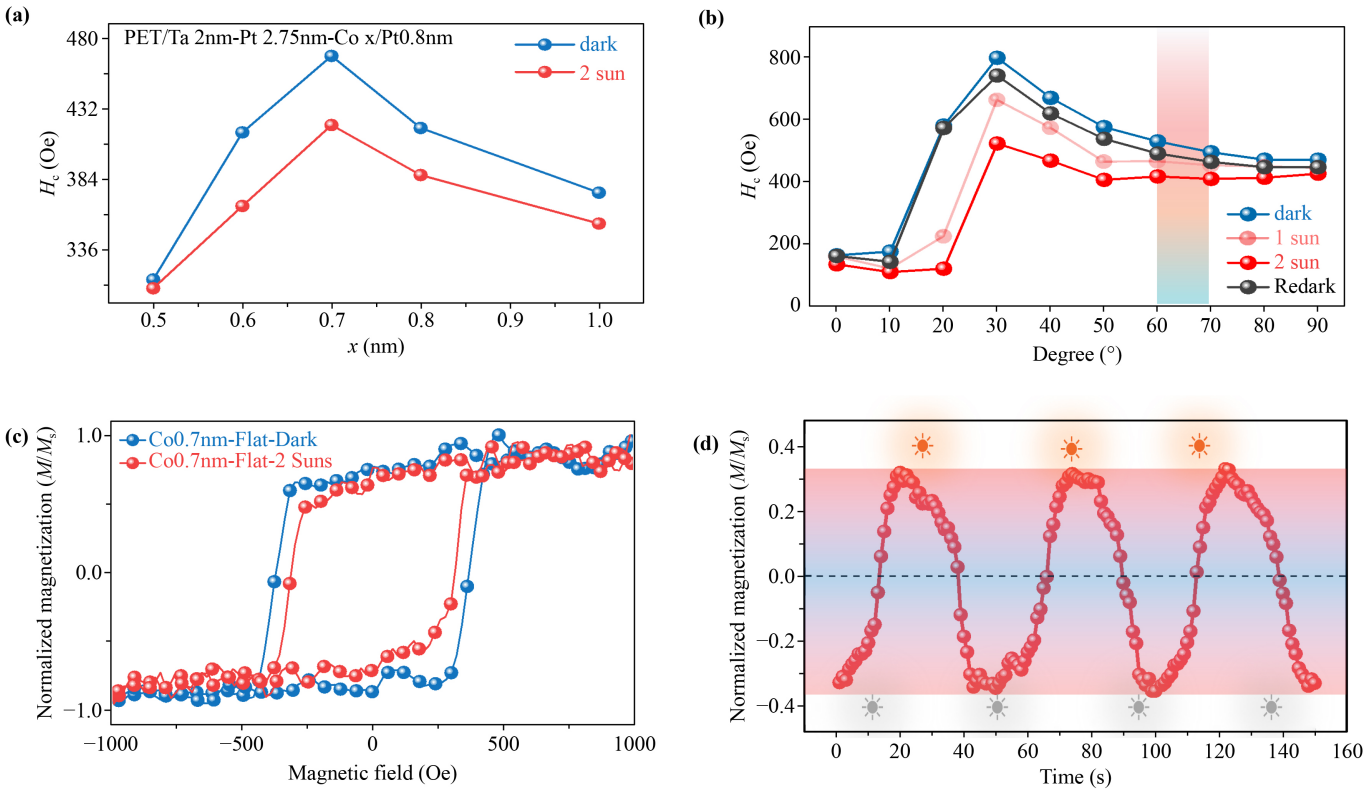

Fig. 2 The sunlight control of magnetic hysteresis loops study in PV/PMA heterostructures on flat conditions. **(a)** The H_c dependence of the thickness of the Co layer with (red) and without (blue) sunlight illumination. **(b)** The angular dependence of H_c without sunlight illumination (dark, red), under 1 sun (blue), 2 suns illumination (green), and back to dark conditions (re-dark, black). **(c)** The VSM measured magnetic hysteresis loops with (red) and without (blue) sunlight illuminations on flat conditions. **(d)** The magnetization switching with sunlight illumination turn on and off with magnetic bias assistance.

purchased from 1-Material Chemscitech Inc. (Canada). Lastly, a 3 nm Pt layer was deposited as the top electrode.

2.2 *In situ* magnetic property measurement

The *in situ* VSM measurement was taken with a LakeShore 7404 VSM system. The device was attached to a rotator, which shows the angle between the film plane and the applied magnetic field. The frequency of the TE 011 mode microwave was 9200 MHz. Devices were illuminated under AM 1.5 G (100 mW·cm^{-2}) using a PL-XQ500W Xenon lamp solar simulator. The intensity of sunlight illumination is 200 mW·cm^{-2} (2 suns).

3 Results and discussion

Figure 1(c) shows the magnetic hysteresis (M–H) loops measured by vibrating sample measurements (VSM) of OPV/Pt (0.8 nm)/Co (0.7 nm)/Pt (2.75 nm) PMA heterostructure on the PET substrate at the flat conditions. Both in-plane (IP) and out-of-plane (OP) M–H loops are measured to identify a typical PMA, and the OP M–H loop is along the magnetic easy-axis compared to the IP M–H loop. The Co thickness is carefully optimized to establish appropriate PMA strength for sunlight tuning, where too strong PMA is challenging to be manipulated.

Under sunlight illumination up to 200 mW·cm^{-2}, referring to two standard suns, the H_c of the PMA M–H loops is significantly reduced, as demonstrated in Figs. 2(a)–(c). In Fig. 2(a), the Co thickness dependence of H_c change before and after sunlight illumination was systematically measured, and the maximum H_c change from 470 Oe (dark) to 410 Oe (2 suns illumination) appears at 0.7 nm Co thickness. When Co thickness goes lower to 0.5 nm, the PMA gradually disappears due to inevitable substrate roughness, decreasing H_c tunability. The substrate roughness is illustrated in Fig. S3. It demonstrates that higher surface roughness prevents the formation of thin-film heterostructure of PMA effect [Pt (0.8 nm)/Co (0.7 nm)/Pt (2.75 nm)]. As the Co thickness increases to 1 nm, the PMA strength enhances accordingly, resulting in a relatively small sunlight tunability. It attributes to the photo-induced electrons doping so that the Fermi level can be shifted to enchance the in-plane Rashba field within Co/Pt interfaces, resulting in weakened PMA according to our previous work [40].

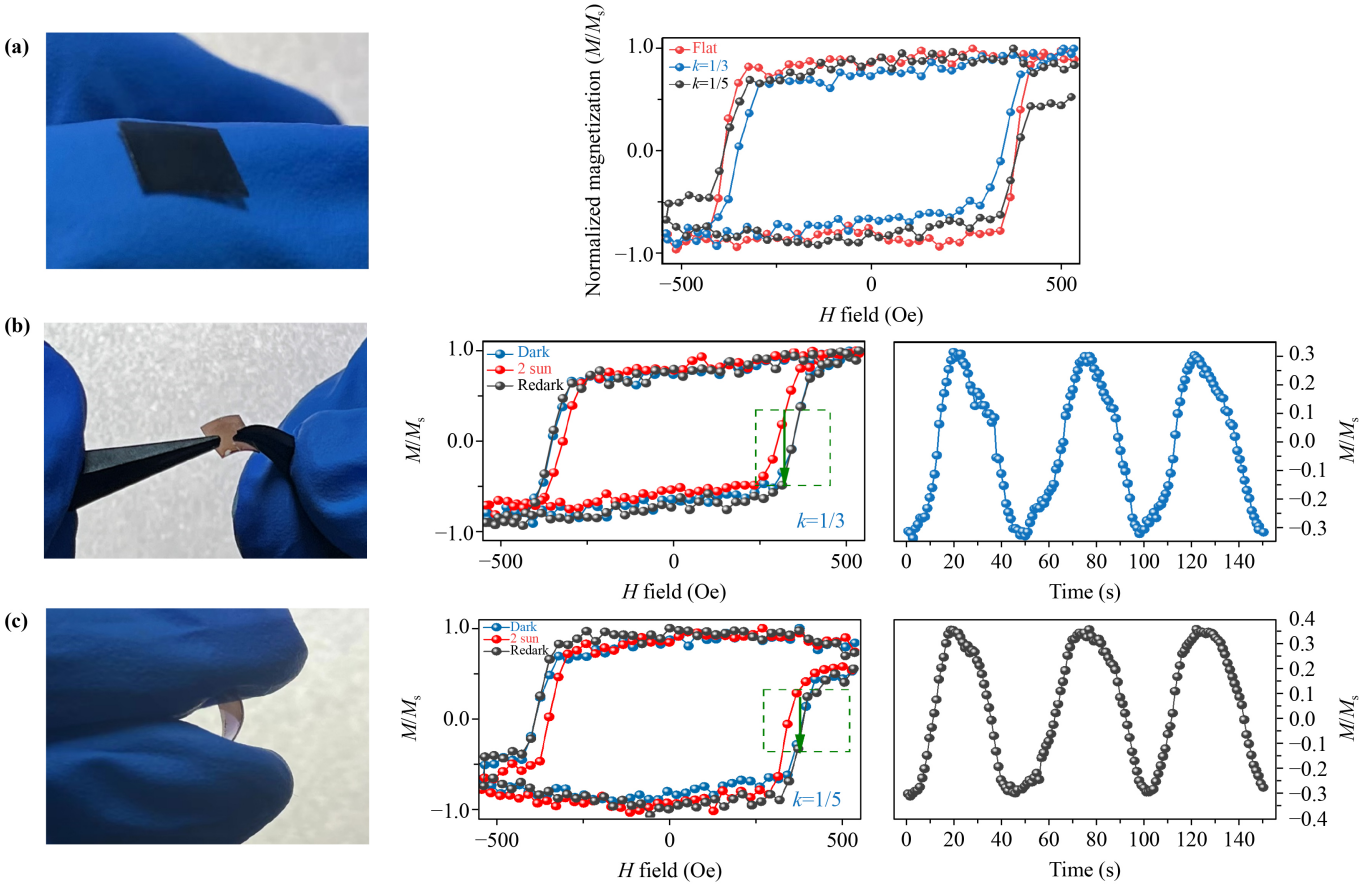

Fig. 3 The sunlight control of PMA on flexible substrates under different bending conditions. **(a)** The magnetic hysteresis loop test of PV/PMA heterostructure under flat (red), bending (curvature = 1/3, blue; 1/5, black) conditions. **(b)** The magnetic hysteresis loop of PV/PMA heterostructure with and without sunlight illumination (dark, blue; redark, black) at substrate bending curvature 1/3. The magnetization switches by turning the sunlight on and off with magnetic bias. **(c)** The magnetic hysteresis loop of PV/PMA heterostructure with and without sunlight illumination (dark, blue; redark, black) at substrate bending curvature of 1/5. The corresponding sunlight-induced magnetization switches at magnetic bias.

Meanwhile, the angular dependence (the angle θ between the applied magnetic field and device-film-plane) of H_c tunability is also investigated in Figs. 2(b) and S1 (Supporting Information). The maximum sunlight control of H_c tunability (from ~800 Oe to ~500 Oe, referring to 60% tunability) was obtained at θ = 30°. In contrast, at θ = 0° and 90°, the tunability decreases to zero. The large H_c tunability enables a sunlight control of deterministic magnetization switching with a slight magnetic bias, as in our previous work in PMA/P–N Si heterojunctions, where a near 180-degree sunlight tunable magnetization switching was achieved. However, the optimized magnetic field angle for magnetization switching in this experiment is not 30° because the M–H loop is highly tilted [40]. Here, we fixed the θ = 70°, where the tunability and the squareness of the M–H loop are balanced. As shown in Fig. 2(c), the M– H loop becomes compact, with H_c decreasing from 470 Oe (blue) to 410 Oe (red) under 2 suns illumination. While applying a magnetic bias of ~440 Oe, the magnetization can be switched from negative (dark) to positive (light) with good reversibility under sunlight, as presented in Fig. 2(d). Indeed, the PMA and magnetization switching angle tunability in the flexible substrate is not comparable to those on P–N Si. The reasons are attributed to the relatively large roughness and non-compensable residual stress of flexible PET substrates and the lower photoelectron generation rate of OPV than that of P–N Si.

In order to examine the stability of flexible PV/PMA devices under various strain/stress conditions, the M–H loops under flat and bending (with curvatures of 1/3 and 1/5, respectively) conditions are tested, as shown in Fig. 3(a). By bending the film, the loops of PMA effect can be altered due to the inverse magnetostriction effect. It attributes to the negative magnetostriction coefficient of cobalt [30]. The M–H loops vary as the bending curvature changes from flat to 1/3 and 1/5, respectively, proving a strain-induced magnetic anisotropy change. While the bending curvature maintains 1/3, as represented in Fig. 3(b), the H_c of the M–H loop still reversibly decreases under 2 suns illumination, which means that the M–H loop returns to its original after withdrawing

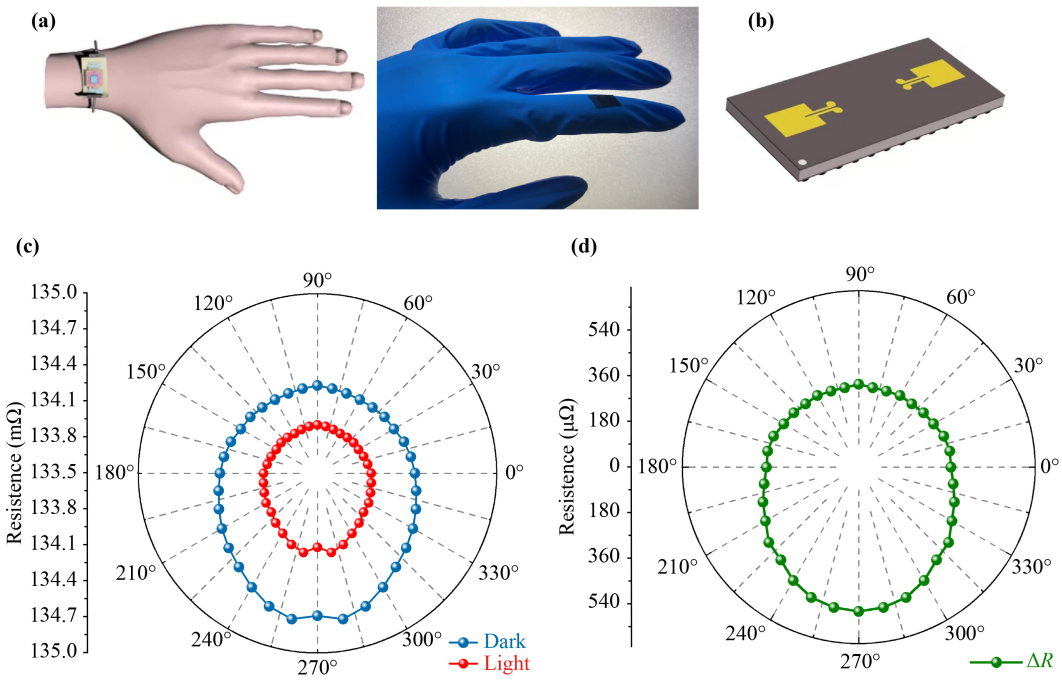

Fig. 4 The prototype device of the solar-driven spintronic sensor. **(a)** The schematic and actual photo of the solar-driven rotating sensor on a human hand. **(b)** The schematic of electrodes on the PV/PMA heterostructures. **(c)** The angular dependence of voltage output with (blue) and without (red) sunlight illuminations. **(d)** The angular dependence of voltage output difference between light and dark conditions.

the sunlight. By applying ~420 Oe magnetic bias, the magnetization also switches from negative (dark) to positive (light) back and forth as it is under flat conditions [Fig. 2(d)]. Similarly, at the bending curvature of 1/5 [Fig. 3(c)], both M–H loops, H_c, and magnetization switching at a fixed magnetic bias (~380 Oe) are tunable by sunlight with good reversibility, showing that the solar-driven PMA change, as well as magnetization switching, can be applied to flexible substrates with complex bending conditions for real flexible spintronic applications.

The angular dependence of sunlight-tunable magnetoresistance (M_R) is also measured for real applications that the input and output signals are voltage or current. Figure 4(a) shows the schematic and actual photo of the prototype solar-driven spintronic sensor on a human hand for potential wearable electronics/spintronics. Figure 4(b) represents the two-point electrodes on the device for magnetoresistance testing. By rotating the applied external magnetic field 360° and fixing the magnetic field to 500 Oe, the angular M_R with (red) and without (blue) sunlight are summarized, showing a ~0.55 mΩ M_R change at its maximum angle of 270° [Fig. 4(c)]. Figure 4(d) demonstrates the angular dependence of M_R under sunlight in a ~0.3–0.55 mΩ range. Although the M_R change is relatively small, solar-driven flexible spintronics is indeed feasible, and it will keep improving rapidly by optimizing the OPV efficiency, ferromagnetic heterostructure, and substrate selections.

4 Conclusion

In conclusion, we realized a solar-tunable PMA heterostructure flexible substrates, where the H_c can be effectively tuned by sunlight illumination from 470 Oe to 410 Oe with 14.6% tunability. With a slight magnetic bias of 320 Oe, magnetization orientation can be manipulated back and forth by turning the sunlight on and off, creating a "0" and "1" information unit. Under pending conditions ($k = 1/3, 1/5$), the PV/PMA devices maintain effective sunlight tunability with strain/stress stability. Lastly, we tested a sunlight-tunable flexible sensor based on our structure, which demonstrated a good angular-dependent sunlight response and paved the way toward solar-driven flexible spintronics in the upcoming years.

Declarations The authors declare that they have no competing interests and there are no conflicts.

Electronic supplementary materials The online version contains supplementary material available at https://doi.org/10.1007/s11467-023-1377-0 and https://journal.hep.com.cn/fop/EN/10.1007/s11467-023-1377-0.

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