Supporting Information

Photovoltaic Control of Ferromagnetism for Flexible Spintronics

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1.BHJ Chemical Structure.

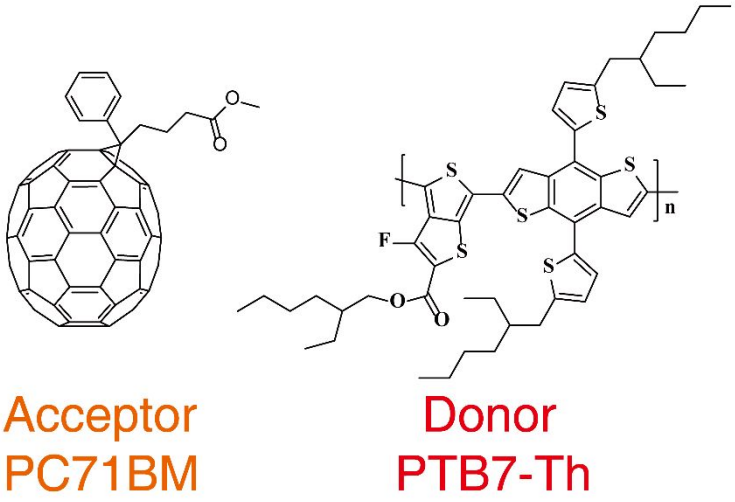

Figure S1 The chemical structure of the donor (PTB7-Th) and acceptor ($PC_{71}BM$) of the body heterostructure junction (BHJ).

2. Elimination of the Temperature Effect

In our experiment, the lab room temperature was 21℃. The light irradiation induced heating was estimated via observing the temperature change of the bottom blackened Kerosene thermometer, which was illuminated by the specific intensity of white light. The 150 mW/cm^{-2} white light could heat the thermometer to 46℃, which we believe is the maximum temperature the light heating could induce. Then the magnetic anisotropy change under 46℃ atmosphere was characterized as demonstrated in Figure S2, and it showed that the heating caused magnetic anisotropy change was not comparable with the illumination induced magnetic anisotropy change. The 1.5-sun white illumination could induce $\Delta H_{r,ip} = 75.4\ Oe$ and $\Delta H_{r,op} = -495.9\ Oe$, corresponding to a $\Delta H_a = -571.3\ Oe$. And the 46℃ environment could induce $\Delta H_{r,ip} = -18.9\ Oe$ and $\Delta H_{r,op} = -37.6\ Oe$, corresponding to a $\Delta H_a = -18.7\ Oe$.

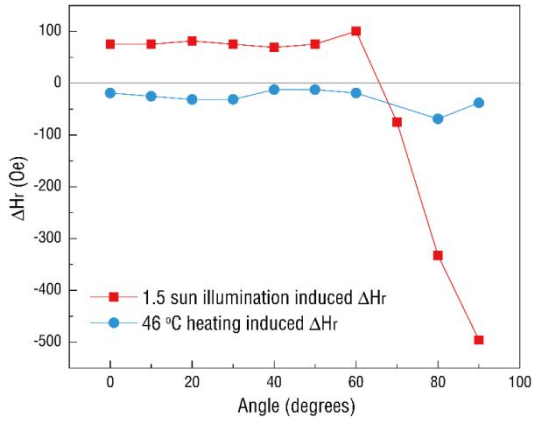

Figure S2 Angular dependence of FMR field variation induced by the illumination effect as well as the heating effect ESR measurement.

3. The interplay between the photovoltaic and strain effects

We measured the photovoltaic voltage as a function of the different radius of curvature under certain simulated sunlight intensity of 100 mW/cm^{2,} as shown in Figure S3. Firstly, 1.5 mm wide and 1 nm thick Co strip was deposited on to the ~1.5 cm x 1.5 cm Kapton substrate with DC sputtering. Then ~0.5 cm x 0.5 cm photovoltaic layer was spin-coated onto the Kapton, and the two ends of the Co stripe were uncovered. Then, 1.5 mm wide and 3 nm thick Pt strip was deposited on to the photovoltaic layer vertical to the Co strip similar to the structure, as shown in the photo in Figure S9. In the voltage measurement, the positive probe from the Keithley 2182 Nano voltage meter was connected to the Pt, and the negative probe was connected to the Co. We can observe that the open voltage is not dependent on the bending radius, which implies that there is no obvious interplay between the photovoltaic and strain effects.

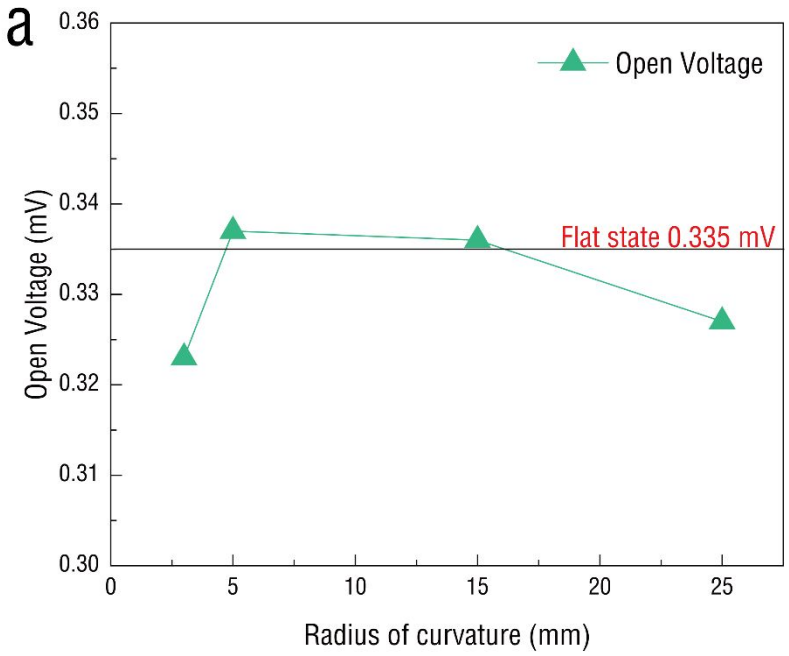

Figure S3 Photovoltaic voltage as a function of the different radius of curvature under certain simulated sunlight intensity of 100 mW/cm^2.

4. Reversibility Test for the Photovoltaic Magnetic Anisotropy Modulation

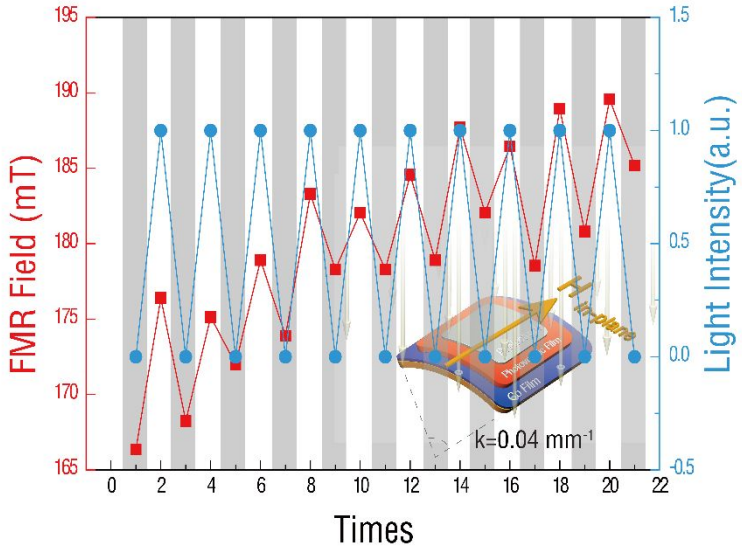

Figure S4 The reversibility of the photovoltaic excited H_r of the k=0.4 mm^{-1} sample along the out-of-plane direction (red-square line) in response to the light switching, and the inset at right corner illustrats the geometry of the measurement. The blue-circle line stands for the alternating light intensity between dark state and 1.0 sun state.

5.ESR Wave Shape of the as grown Sample

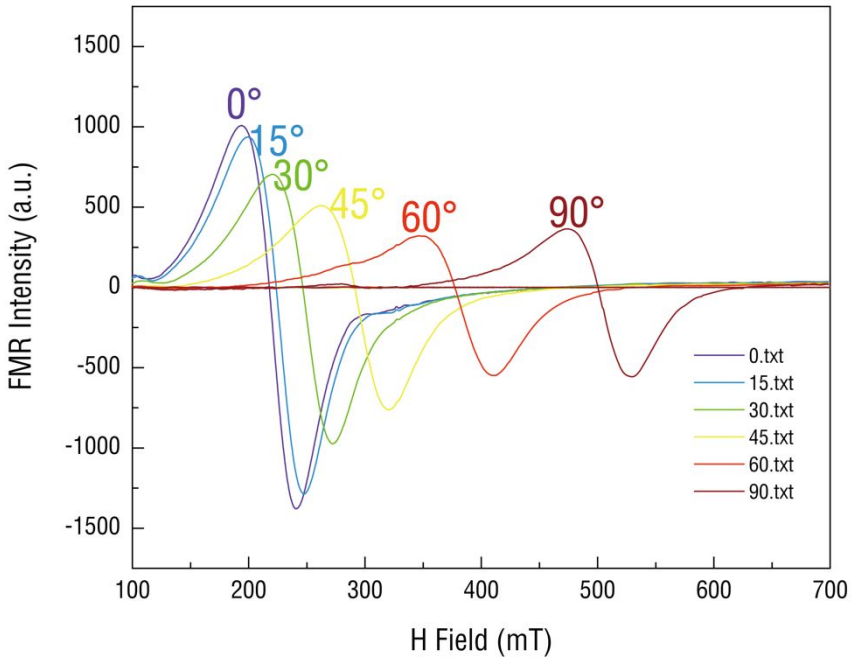

Figure S5 The FMR spectrum at different angles of the as-grown Ta(4 nm)/Co(1 nm) sample. The geometry of the sample and the test condition was illustrated in Figure 1(a).

6. Work Function of the electrodes, HUMO and LUMO of the OPV layer

Considering the variation of the work function of the electrodes at the thin-film state and the fluctuation of the HUMO and LUMO value in this device, the energy number used in Figure 5 was carefully measured as shown in Figure S5 and S6. The working function Φ was based on the following equation, $\Phi - h\upsilon = E_{Fermi} - E_{Cutoff}$, where $h\upsilon$ is the source energy, E_{cutoff} and E_{Fermi} stand for the binding energies of the secondary electron cutoff and the Fermi level, respectively.[1] As shown in Figure S6, the E_{cutoff} and E_{Fermi} were determined with the 1st derivative of the kinetic energy in order to find the lower and higher energy edge of the spectrum, respectively.

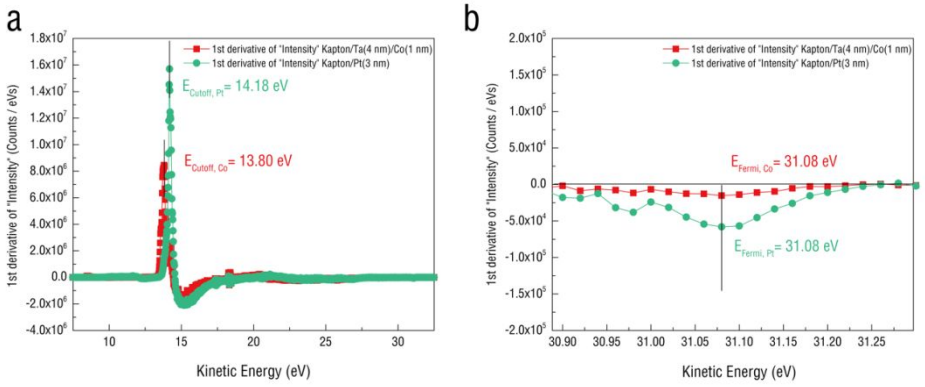

Figure S6 First Derivative of the Work function Φ of the as-grown Co (1 nm) and Pt (3 nm) electrodes with Ultraviolet Photoelectron Spectroscopy (UPS) method

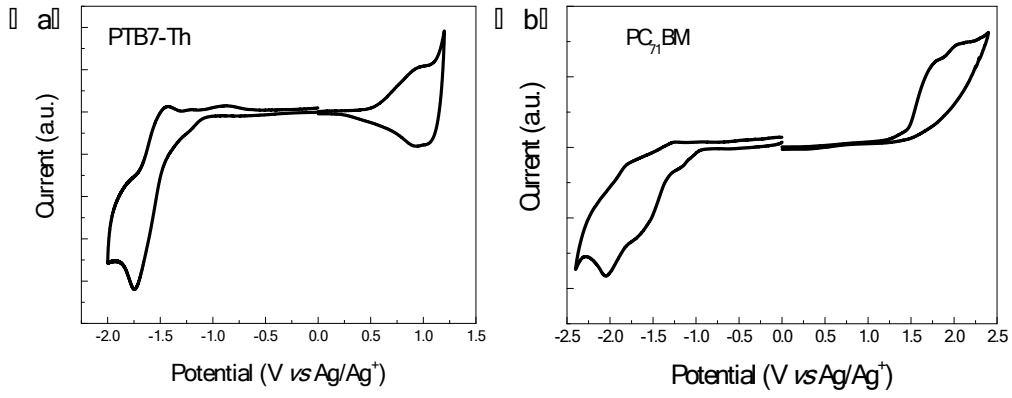

Figure S7. Electrochemical cyclic voltammetry curve of the (a) PTB7-Th film;(b) $PC_{71}BM$ film measured in 0.1 mol L^{-1} Bu_4NPF_6 acetonitrile solutions.

7. Transient Absorption Test For the Mechanism Discussion

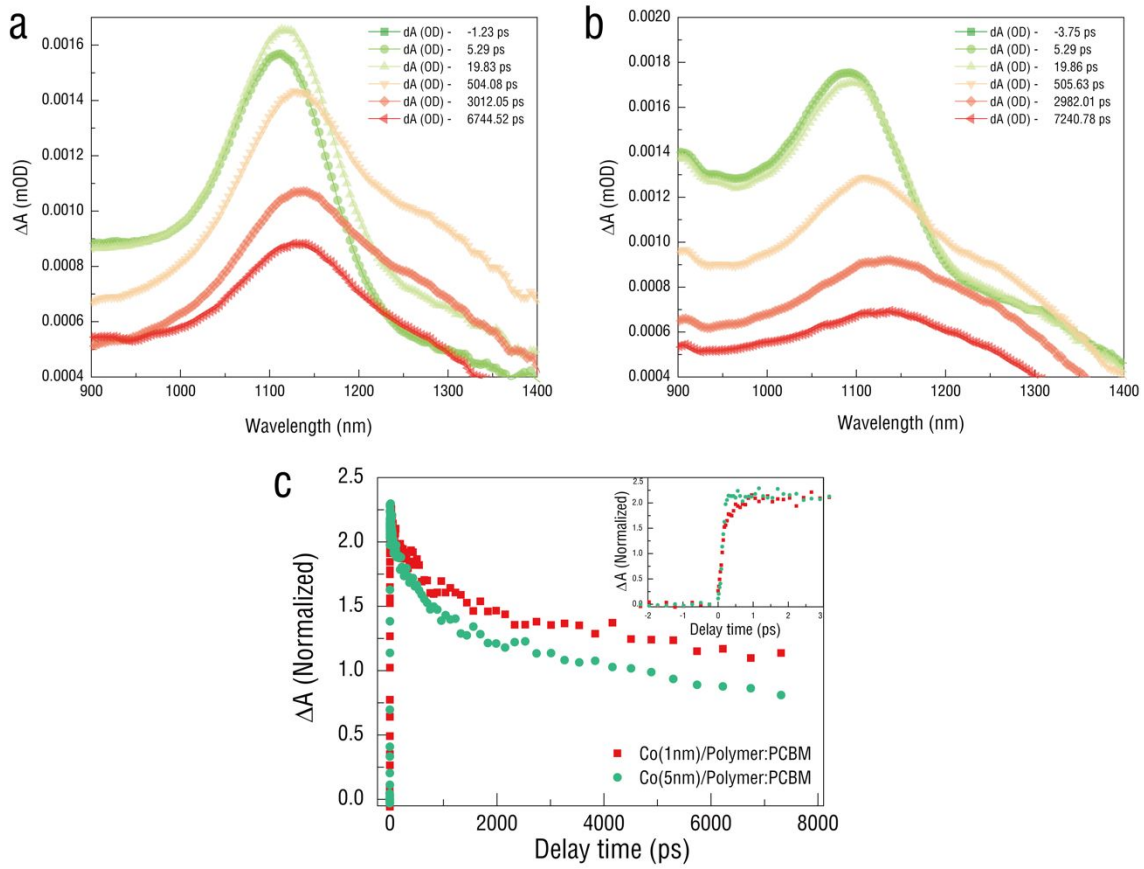

Figure S8 Transient absorption (TA) spectra of PTB7-Th: $PC_{71}BM$ in different media: (a) Kapton/Ta/Co (1 nm)/Photovoltaic layer and (b) Kapton/Ta/Co (5 nm)/Photovoltaic layer at the indicated delay times. The excitation wavelength is 600 nm, with a power of 7 μJ/cm^2/pulse. (c) The normalized absorption of the 1150 nm light as a function of the response time. The inset shows a zoom-in view in the 0~3 ps delay time scale.

The exciton separation was also tested in this flexible heterostructure by the transient absorption spectrum peak at 1100 nm, as demonstrated in Figure S8(a). It is attributed to the formation of a charge-separated state in PTB7-Th: $PC_{71}BM$.[2, 3] Figure S8 gave out the normalized transient absorption as a function of the delay time at 1150 nm wavelength, and the instinct increase within the 2 ps after the illumination start represented the photon excitation of the organic semiconductor $PC_{71}BM$: PTB7-Th.[2]

8. Measurement for the Voltage Difference between the Pt and Co Electrode under Visible Light Illumination.

Firstly, 2 mm wide and 3 nm thick Co strip was deposited on to the ~1.5 cm x 1.5 cm Kapton substrate with DC sputtering. Then ~1 cm x 0.8 cm photovoltaic layer was spin-coated onto the Kapton, and the two ends of the Co stripe were uncovered. Then, 2 mm wide and 3 nm thick Pt strip was deposited on to the photovoltaic layer vertical to the Co strip as shown in the photo in Figure S9. In the voltage measurement, the positive probe from the Keithley 2182 Nano voltage meter was connected to the Pt and the negative probe was connected to the Co.

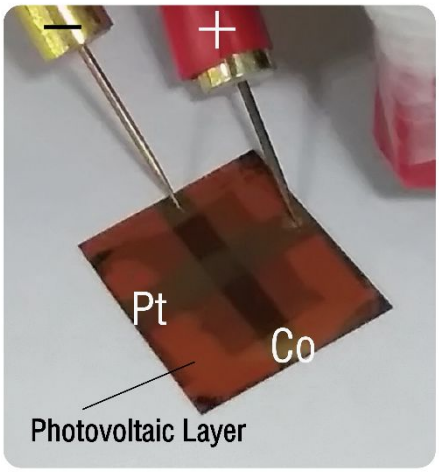

Figure S9 Photo of the geometry for the photovoltaic measurement.

Reference

(1) Song, W. and M. Yoshitake, A Work Function Study of Ultra-Thin Alumina Formation on Nial (1 1 0) Surface. *Appl Surf Sci*, **2005**, 251, 14-18.

(2) Rolczynski, B.S.; J.M. Szarko; H.J. Son; Y. Liang; L. Yu; and L.X. Chen, Ultrafast Intramolecular Exciton Splitting Dynamics in Isolated Low-Band-Gap Polymers and Their Implication in Photovoltaic Materials Design. *J Am Chem Soc*, **2012**, 134, 4142-4152.

(3) Szarko, J.M.; B.S. Rolczynski; S.J. Lou; T. Xu; J. Strzalka; T.J. Marks; L. Yu; and L.X. Chen, Photovoltaic Function and Exciton/Charge Transfer Dynamics in a Highly Efficient Semiconducting Copolymer. *Adv Funct Mater*, **2014**, 24, 10-26.