

## Electronic Supplementary Information

### **Large Magnetoresistance and Spin-Polarized Photocurrent in $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3(\text{Co})/\text{quaterthiophene}/\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ Organic Magnetic Tunnel Junctions**

Xuefei Han<sup>1</sup>, Wenbo Mi<sup>1,\*</sup>, Xiaocha Wang<sup>2,\*</sup>

*<sup>1</sup>Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparation Technology,*

*School of Science, Tianjin University, Tianjin 300354, China*

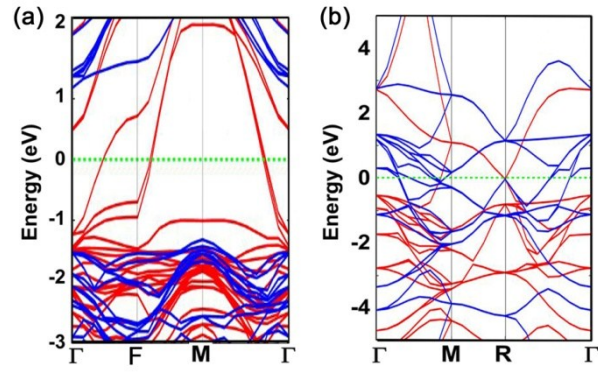
*<sup>2</sup>School of Electrical and Electronic Engineering, Tianjin University of Technology, Tianjin 300384,*

*China*

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\*Author to whom all correspondence should be addressed.

E-mail: [miwenbo@tju.edu.cn](mailto:miwenbo@tju.edu.cn) and [wangxc@email.tjut.edu.cn](mailto:wangxc@email.tjut.edu.cn)



**Fig. S1** Band structure of bulk  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  (a) and Co (b) calculated by Nanodcal package. The red and blue lines distinguish spin-up and spin-down band structure, respectively.

The spin-dependent current was further obtained using the Landauer-Büttiker formula<sup>1</sup>

$$I_{\sigma} = \frac{e}{h} \int_{\mu_L}^{\mu_R} T_{\sigma}(E, V) [f(E - \mu_R) - f(E - \mu_L)] dE, \quad (1)$$

where  $e$  is the electron charge,  $h$  is Planck's constant, and  $T_{\sigma}(E, V)$  is the spin-dependent ( $\sigma = \uparrow, \downarrow$ ) transmission coefficient at a bias voltage  $V = V_R - V_L$ .  $f(E - \mu_{L(R)})$  is Fermi-Dirac distribution function related to the chemical potential  $\mu_{L(R)}$  of the left(right) electrode.

The spin injection efficiency (SIE) and tunneling magnetoresistance (TMR) at a voltage are defined as

$$\text{SIE} = \frac{I_{\uparrow}(V) - I_{\downarrow}(V)}{I_{\uparrow}(V) + I_{\downarrow}(V)}, \quad (2)$$

$$\text{TMR} = \frac{I_{\text{PC}}(V) - I_{\text{APC}}(V)}{I_{\text{APC}}(V)}, \quad (3)$$

where  $I_{\text{PC}}$  and  $I_{\text{APC}}$  are the total currents in PC and APC, respectively. At the equilibrium state, SIE and TMR are calculated from the electron transmittance coefficient ( $T(E_F)$ ) at Fermi level.

For linearly polarized light, the polarization vector can be defined as  $\mathbf{e} = \cos\theta \mathbf{e}_1 + \sin\theta \mathbf{e}_2$ , where  $\theta$  is the angle between the polarization direction and the vector  $\mathbf{e}_1$ . The photocurrent moving into the left electrode is defined as follows<sup>2, 3</sup>

$$J_L^{(\text{ph})} = \frac{ie}{h} \int (\cos^2 \theta \text{Tr} \{ \Gamma_L [G_1^{<(\text{ph})} + f_L (G_1^{>(\text{ph})} - G_1^{<(\text{ph})})] \})$$

$$\begin{aligned}
& + \sin^2 \theta \text{Tr}\{\Gamma_L[G_2^{<(\text{ph})} + f_L(G_2^{>(\text{ph})} - G_2^{<(\text{ph})})]\} \\
& + \frac{\sin(2\theta)}{2} \text{Tr}\{\Gamma_L[G_3^{<(\text{ph})} + f_L(G_3^{>(\text{ph})} - G_3^{<(\text{ph})})]\}dE,
\end{aligned} \tag{4}$$

where

$$\begin{aligned}
G_1^{>/<(\text{ph})} &= \sum_{\alpha, \beta=x, y, z} C_0 N G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a, \\
G_2^{>/<(\text{ph})} &= \sum_{\alpha, \beta=x, y, z} C_0 N G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a, \\
G_3^{>/<(\text{ph})} &= \sum_{\alpha, \beta=x, y, z} C_0 N (G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a + G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a).
\end{aligned} \tag{5}$$

The polarization vector of elliptically polarized light is  $\mathbf{e} = \cos\phi \mathbf{e}_1 + i \sin\phi \mathbf{e}_2$ , where  $\phi$  determines its helicity.  $\phi = \pm 45^\circ$  corresponds to right- and left-handed circularly polarized light, then the photocurrent is described as

$$\begin{aligned}
J_L^{(\text{ph})} &= \frac{ie}{2h} \int (\text{Tr}\{\Gamma_L[G_1^{<(\text{ph})} + f_L(G_1^{>(\text{ph})} - G_1^{<(\text{ph})})]\} \\
& + \text{Tr}\{\Gamma_L[G_2^{<(\text{ph})} + f_L(G_2^{>(\text{ph})} - G_2^{<(\text{ph})})]\} \\
& \pm \text{Tr}\{\Gamma_L[G_3^{<(\text{ph})} + f_L(G_3^{>(\text{ph})} - G_3^{<(\text{ph})})]\})dE,
\end{aligned} \tag{6}$$

where  $G_{1,2}^{>/<(\text{ph})}$  is the same as the linearly polarized case and

$$G_3^{>/<(\text{ph})} = \pm i \sum_{\alpha, \beta=x, y, z} C_0 N (G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a - G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a). \tag{7}$$

In the above equations,  $C_0 = (e/m_0)^2 (\hbar \sqrt{\mu_r \epsilon_r} / 2N\omega \epsilon c) I_\omega$ , where  $m_0$  is the bare electron mass,  $I_\omega$  is the photon flux defined as the number of photons per unit time per unit area,  $\omega$  is the frequency and  $c$  is the speed of light,  $\mu_r$  is the relative magnetic susceptibility,  $\epsilon_r$  is the relative dielectric constant,  $\epsilon$  is the dielectric constant, and  $N$  is the number of photons.  $G_0^{r/a}$  are the retarded and advanced

Green's functions without photons, respectively. Meanwhile,  $G_0^{>/<}$  are greater and lesser Green's function without photons, respectively.  $p_{x,y,z}$  is the Cartesian component of the electron momentum, and  $e_{1/2x,y,z}$  is the Cartesian component of the unit vector  $\mathbf{e}_{1/2}$ , which characterizes the polarization of the light. The normalized photocurrent is written as

$$R = \frac{J_L^{(\text{ph})}}{eI_\omega}. \quad (8)$$

Note that the unit of  $R$  is  $a_0^2/\text{photon}$  where  $a_0$  is the Bohr radius.

## References

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