

**Solution-processed organic spin–charge converter**

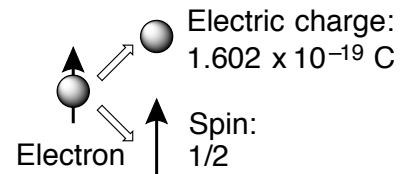
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**1. Introduction**

**1.1. Spintronics**

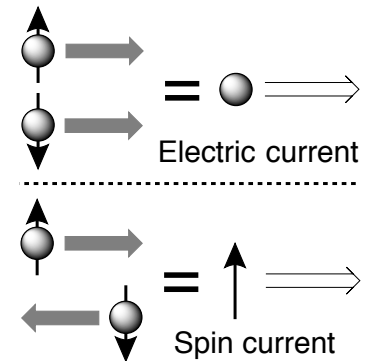
- Electron has two types of properties, 1: electric charge, 2: spin (*Figure 1*).
- Utility of spin in addition to electrical charge will opens up new types of low-energy-consuming nanoelectronic devices. (e.g. Magnetic RAM, Spin transistor, High sensitivity magnetic sensor)



**Figure 1.** Two properties of electron.

**1.2. Organic materials on spintronics**

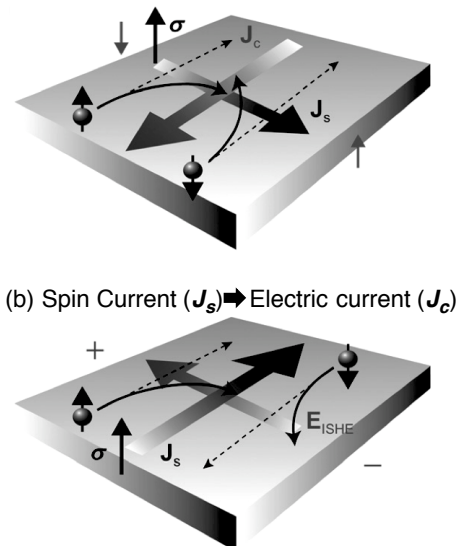
- Organic materials have advantages over inorganic materials such as flexibility, large-area processability and low-cost manufacturing.
- Also, small spin–orbit coupling of organic material causes a long spin lifetime.
- In order to use spin of organic materials, a method to convert spin information to an electric signal is indispensable.



**Figure 2.** Two types of currents.  
(a) Electric current ( $J_c$ )  $\rightarrow$  Spin Current ( $J_s$ )

**1.3. Conversion between electric charge and spin information**

- Electric current is the entity to store electronic information.
- Spin current instead, is the entity to store spin information (*Figure 2*).
- Spin current is generated in following mechanism (*Figure 3a*):
  - When certain voltage is applied, a flow of electrons,  $J_c$ , is generated.
  - Due to spin polarization  $\sigma$ , electrons gain force perpendicular to both  $J_c$  and  $\sigma$ .
  - When spin polarization  $\sigma$  is aligned in a certain direction due to spin–orbit coupling, spin current  $J_s$  is generated. This is called spin Hall effect, SHE.
- In the opposite case, where spin current  $J_s$  is generated, electric current  $J_c$  is induced (*Figure 3b*). This is called inverse spin Hall effect, ISHE. ( $E_{ISHE} \parallel J_s \times \sigma$ )



**Figure 3.** SHE and ISHE.

#### 1.4. Difficulty on detection of ISHE in organic materials

- Detection of ISHE in inorganic materials is relatively easy due to their stronger spin-orbit coupling. (cf. Pt)
- Detection of small voltage difference is required for observation of ISHE in organic materials, since they have smaller spin-orbit coupling.
- Any possible noise that comes from injection of spin current should be avoided.

#### 1.5. Spin pump<sup>1</sup>: a strong method for spin injection in various materials<sup>2</sup>

- In this method, no voltage is applied to active material. Thus, electric noise is small.

- Spin pump from ferromagnetic metal towards non-magnetic material is performed in following procedure (Figure 4):

- Magnetic field  $H$  is applied in ferromagnetic metal.
- Microwave irradiation causes precession of magnetic moment  $M(t)$ . This situation is similar to NMR, in which magnetic moment of a nucleus is considered instead of magnetization that comes from whole magnetic moment of a material.
- The precession movement injects spin current  $J_s$  into non-magnetic material.

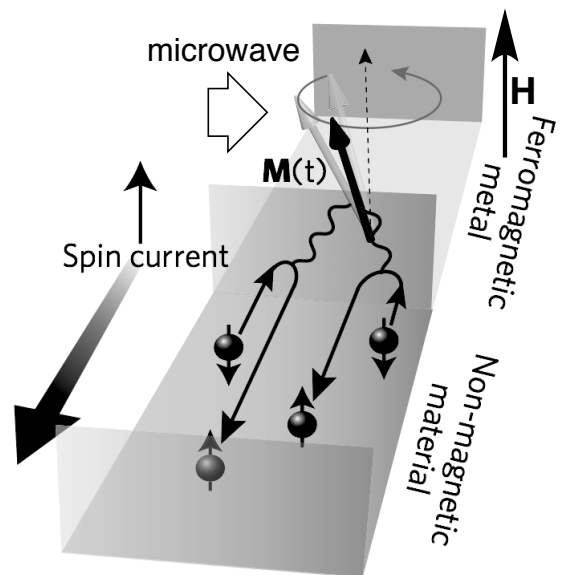


Figure 4. Spin pump method.

#### 1.6. This work

- Detection of ISHE in an organic material for the first time.
- The use of *magnetic insulator*,  $Y_3Fe_5O_{12}$ ,<sup>3</sup> for injection of spin current, instead of conventional ferromagnetic metal, further diminishes noise in the organic material.

## 2. Methods

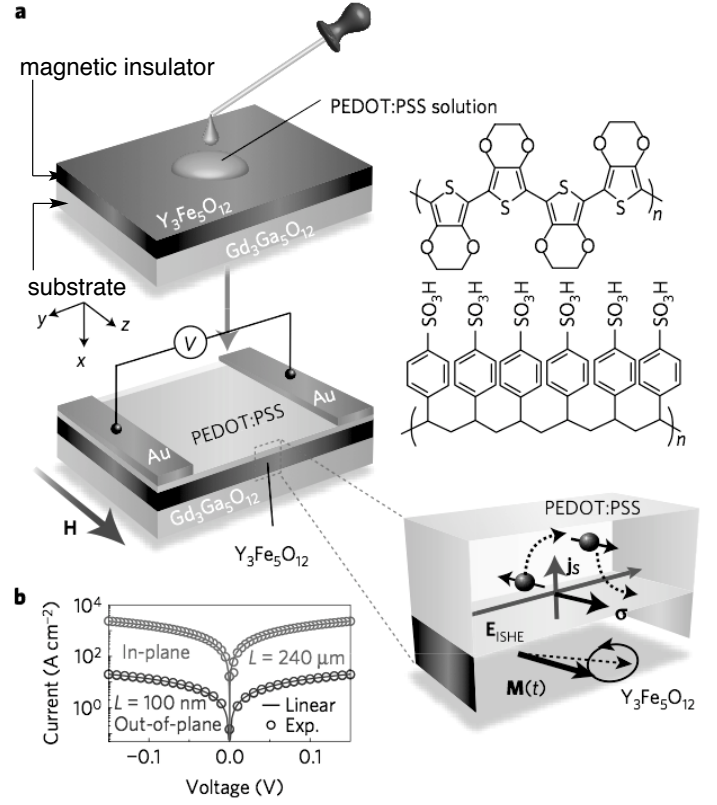
### 2.1 Device configuration

- $Gd_3Ga_5O_{12}$  (substrate)/ $Y_3Fe_5O_{12}$  (5  $\mu m$ )/PEDOT:PSS (80 nm)/Au (Figure 5a)
- $Y_3Fe_5O_{12}$  was grown on  $Gd_3Ga_5O_{12}$  substrate by liquid-phase epitaxy.

- The authors used PEDOT:PSS, which is a polymer conductor with high in-plane conductivity of around  $10^3 \text{ S cm}^{-1}$  with large anisotropy (Figure 5b).

## 2.2. Pumping spin current in PEDOT:PSS

- A magnetic field  $\mathbf{H}$  was set to the direction of  $z$  axis.
- Microwave was set to the direction of  $x$  axis to induce precession of magnetization  $\mathbf{M}(t)$  in  $\text{Y}_3\text{Fe}_5\text{O}_{12}$ .
- Spin current  $\mathbf{J}_s$  is injected into PEDOT:PSS to the direction of  $-x$  axis.



**Figure 5.** (a) Device configuration for measurement of  $V_{\text{ISHE}}$ . (b) Conductance anisotropy of PEDOT:PSS.

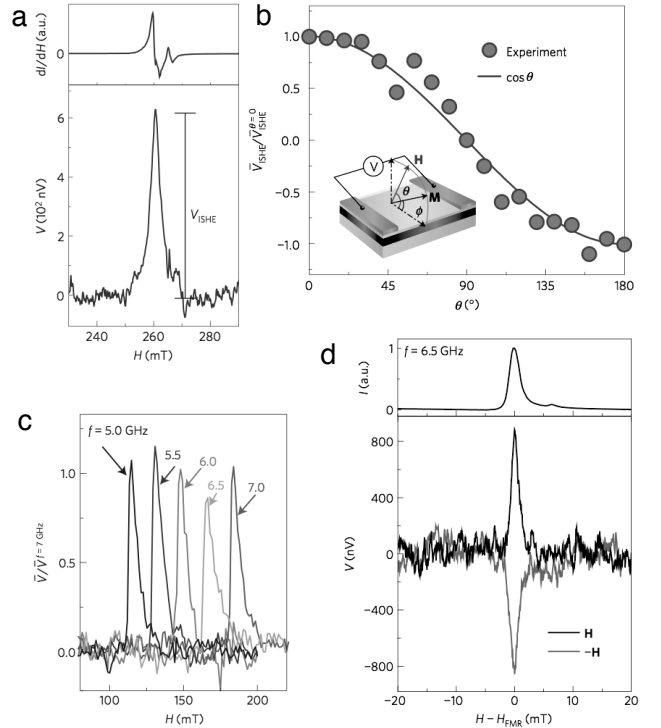
## 3. Results and discussion

### 3.1 Observation of electric voltage difference between two electrodes

- In above conditions, electric voltage difference  $V_{\text{ISHE}}$  was generated (Figure 6a).
- Dependence of  $V_{\text{ISHE}}$  against  $\theta$  is consistent with ISHE symmetry (Figure 6b).<sup>4</sup>
- This electric field is expected to be induced by ISHE, however, several possible reasons should be excluded, such as  $\mathbf{H}$ -dependent and/or independent thermoelectric effect by microwave irradiation of PEDOT:PSS.

### 3.2 Exclusion of other possible reasons

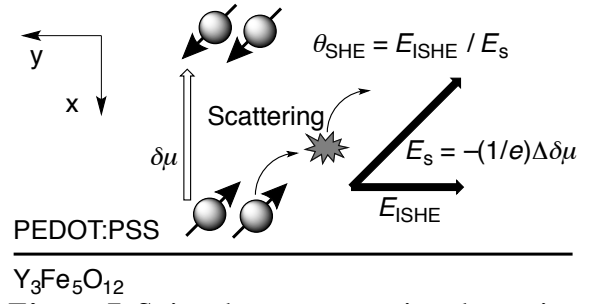
- Absence of  $\mathbf{H}$ -dependent thermoelectric effect was confirmed by checking voltage difference under various magnetic field  $\mathbf{H}$  (Figure 6c).
- Reversing magnetic field  $\mathbf{H}$  caused inversion of electric field without significant change of  $|V_{\text{ISHE}}|$  (Figure 6d), indicating no  $\mathbf{H}$ -independent thermoelectric effect occurred.



**Figure 6.** Electric voltage detection at several magnetic resonance conditions.

### 3.3 Investigation of conversion efficiency

- Spin accumulation at the interface  $\delta\mu_0$  was calculated from Bloch equation for carrier spin.<sup>3</sup> PEDOT:PSS had higher  $\delta\mu_0$  than Pt due to longer spin lifetime (*Figure 7*).
- Spin Hall angle  $\theta_{\text{SHE}}$  of PEDOT:PSS was smaller than Pt due to weaker spin-orbit coupling.
- The large conductivity anisotropy of PEDOT:PSS ( $\sigma_c^y/\sigma_c^x \sim 4 \times 10^5$ ) enhances  $J_c/J_s$  conversion efficiency  $\alpha_{\text{ISHE}}$  to a comparable value to Pt.



**Figure 7.** Spin-charge conversion dynamics.

**Table 1.** Comparison of ISHE parameters.

	$\delta\mu_0$ ( $\hbar[\mathbf{M} \times \partial_t \mathbf{M}]_z$ )	$\theta_{\text{SHE}}$	$\alpha_{\text{ISHE}}$
PEDOT:PSS	$2 \times 10^{-1}$	$10^{-7}$	$4 \times 10^{-2}$
Pt (ref. 4)	$6 \times 10^{-4}$	$4 \times 10^{-2}$	$4 \times 10^{-2}$
PEDOT:PSS/Pt	$\sim 4 \times 10^2$	$\sim 10^{-5}$	$\sim 1$

### 4. Conclusions

- ISHE of organic materials was observed for the first time using PEDOT:PSS.
- Spin-charge conversion efficiency was comparable to Pt case due to canceling between smaller spin-orbit coupling and large conductivity anisotropy.

### 5. Perspective

- The microscopic origin of the spin-charge conversion in organic materials is now open for discussion.
- In-depth theoretical studies of the spin-charge conversion in PEDOT:PSS will be stimulated by the results reported in this work.
- The almost-infinite chemical tunability of organic materials paves the way towards molecular-structure engineering of spin-charge conversion in condensed matter.

### 6. References

- (1) Tserkovnyak, Y.; Brataas, A.; Bauer, G. *Phys. Rev. Lett.* **2002**, *88*, 117601. (2) Ando, K.; Takahashi, S.; Ieda, J.; Kurebayashi, H.; Trypiniotis, T.; Barnes, C. H. W.; Maekawa, S.; Saitoh, E. *Nat. Mater.* **2011**, *10*, 655. (3) Kajiwara, Y.; Harii, K.; Takahashi, S.; Ohe, J.; Uchida, K.; Mizuguchi, M.; Umezawa, H.; Kawai, H.; Ando, K.; Takanashi, K.; Maekawa, S.; Saitoh, E. *Nature* **2010**, *464*, 262. (4) Ando, K.; Takahashi, S.; Ieda, J.; Kajiwara, Y.; Nakayama, H.; Yoshino, T.; Harii, K.; Fujikawa, Y.; Matsuo, M.; Maekawa, S.; Saitoh, E. *J. Appl. Phys.* **2011**, *109*, 103913.