K-space Orientation Dependent of the Spin Texture in SF$_3$PbI$_3$ Perovskite Compounds

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Abstract. The k-space spin textures in the first Brillouin zone play an essential role in determining how spin-polarized currents can be manipulated for spin-orbitronics based devices. Here, achieving unidirectional spin textures called as a persistent spin helix is important to induce very long spin lifetime, which is useful for controlling spin-polarized currents. Using density functional theory (DFT) with fully relativistic calculations, we show that the unidirectional spin textures are achieved in the k-space of bulk SF$_3$PbI$_3$ oriented on [110]-direction. We found that the Rashba parameter strength of the spin-orbit induced spin splitting is sizable, indicating that this material is promising for energy saving spintronics.

Keywords: Spin textures, perovskite, persistent spin helix, spintronics

1. Introduction
The energy saving spintronics is believed to be achievable by using materials having a unique property called persistent spin helix (PSH) that can enhance the spin relaxation time. The PSH materials exhibit unidirectional spin textures induced by the spin-orbit interaction (SOI). For spintronics devices operator, the PSH materials with strong SOI is required. Therefore, finding materials having strong SOI is crucially important. One of the candidates for spintronics application is the halide hybrid organic-inorganic perovskite (HOIP) [1–3].

One example of this material which is currently under extensive research for photovoltaics application is CH$_3$NH$_3$PbI$_3$. It was reported that CH$_3$NH$_3$PbI$_3$ possesses strong SOI, indicated by the existence of Rashba splitting [4]. Another interesting trait from CH$_3$NH$_3$PbI$_3$ is that the dynamic of the organic molecule as a cation in its crystal system can affect the electronics properties [5]. Furthermore, it has been reported that among many HOIP materials, CH$_3$NH$_3$PbI$_3$ and SF$_3$PbI$_3$ exhibited as the most stable against spontaneous decomposition [6]. Based on such materials, SF$_3$PbI$_3$ with the similar crystal structure to CH$_3$NH$_3$PbI$_3$ is a hypothetical material that hasn’t been studied much yet. Therefore, it is important to identify the spintronics functionality of HOIP.

This paper focuses on the SF$_3$PbI$_3$ system and clarify how the dynamic of crystal orientation in k-space can affect the spin textures of SF$_3$PbI$_3$. Interestingly, we performed fully relativistic DFT and found that the unidirectional spin texture can be achieved in k-space of the bulk SF$_3$PbI$_3$ system. It indicates that such system could find PSH state supported by strong SOI.
2. Methods
The hypothetical material SF$_3$PbI$_3$ used in this calculation was based on pseudo-cubic crystal structure from previous work [6] with cation orientation in [111] direction (Figure 1(a)). For the calculation, the only one-unit cell was considered as a bulk system, where the first Brillouin zone is shown in Figure 1 (b). We performed the calculation of electronic structure using DFT with generalized gradient approximation (GGA) [7]. By using a confinement scheme, the wave functions of this calculation were generated based on the linear combination of multiple pseudo-atomic orbitals (LCPAOs) [8,9]. The orbitals used in the calculation were Pb8.0-$s^2p^2d^2f$, I7.0-$s^2p^2d^2f$, S7.0-$s^2p^2d^2$, F6.0-$s^2p^2d^2$ meaning that the cut-off radii for Pb, I, S, and F is 8.0, 7.0, 7.0, and 6.0, respectively. Spin-orbit coupling was also included in the calculation with non-collinear spin polarization and energy cut-off 300 Ry with k-grid of 8×8×8. By utilizing the spin density matrix of the spinor wavefunction, the spin textures can be calculated [10]. Here, two different crystal orientation were considered for [001] and [110]. We compared the results from the same crystal system but with different orientation in k-space.

3. Results and Discussion
The optimization for the lattice parameters of SF$_3$PbI$_3$ with crystal model is shown in Figure 1 (a). It is found that the calculated value (6.5745 Å) is consistent with previous calculation [6]. However, this value is slightly larger than those data as reported in previous DFT calculation [6] using the same crystal structure with the value of 6.546 Å. The band structure calculation was performed with chosen k-path of Γ-R-X-M-Γ on the first Brillouin zone as shown in Figure 1 (b). The electronics band structure and the projected density of states of SF$_3$PbI$_3$ with and without SOI are shown in Figure 2 (a). We found that a big parameter of spin splitting occurred around M and R point.

Figure 2 (b)-(c) shows the highlight spin splitting around R and M point. We estimated the Rashba spin-orbit strength or Rashba parameter by using free electron model defined as $\alpha = 2E_R/k_R$; where $\alpha$ is the parameter Rashba, $E_R$ and $k_R$ are the respective energy splitting and the k-momentum offset. We also found that the calculated result of parameter Rashba in R point is $\alpha_R = 0.85$ eV.A and in M point is $\alpha_M = 1.611$ eV.A. Such results are comparably larger from a previous report [2].

Figure 1. (a) Structure of SF$_3$PbI$_3$ and (b) first Brillouin zone of cubic SF$_3$PbI$_3$
To demonstrate the properties of the spin splitting, we investigate the k-space spin textures in M point. In this case, the spin textures were calculated at 1.5 eV above the valence band maximum (VBM). It was also found that the conduction band minimum (CBM) had a larger spin splitting than those of the VBM. Therefore, in this paper, we focus only on the studies on spin-split at the CBM. Figure 3 shows the change in spin textures with different crystal orientation, namely with orientation of [001] as shown in Figure 2 (a) and [110]. We can see from Figure 2 (a) for the [001] direction that isotropic Rashba splitting appears. The color palette indicates the in-plane and out-of-plane spin polarization components. However, these spin directions are not unidirectional which are the characteristic of pure Rashba splitting component. Interestingly, we fund anisotropic spin splitting for the case of [110] crystal orientation, exhibited fully out of plane (unidirectional) spin orientation. These typical spin textures indicate that the PSH is achieved. If we compare to previous work, the different crystal system [11], such system provided an extremely long spin lifetime. This characteristic is crucial for developing spin-polarized current for energy saving based spintronics devices.
Figure 3. K-space spin texture of SF$_3$PbI$_3$ around M point with (a) orientation in [001] direction, and (b) in [110] direction.

4. Conclusion
In summary, by using fully relativistic DFT calculation, we found a strong SOI presence in bulk SF$_3$PbI$_3$. Spin textures investigation indicates the existence of PSH in [110]-direction which will induce long spin lifetime. We also found that the k-space spin textures for this system are dependent on the crystal orientation. Although still a theoretical material, SF$_3$PbI$_3$ have excellent potential for future energy saving spintronics devices.

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