Persistent spin helix on a wurtzite ZnO(1010) surface: First-principles density-functional study

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The persistent spin helix (PSH) that has been widely and exclusively studied in zinc-blende structures is revealed for the first time on the surface of a wurtzite structure. Through first principles calculations of the surface of ZnO(1010), a quasi-one-dimensional orientation of the spin textures is identified. Furthermore, the wavelength of this particular PSH is smaller than that observed in various zinc-blende quantum well structures, thus indicating that wurtzite-structured surfaces are suitable for spintronics applications. © 2015 The Japan Society of Applied Physics

Spin-orbit coupled systems have attracted considerable scientific interest over recent years, as they allow for the manipulation of electron spin.1 This spin–orbit coupling (SOC) appears to play an important role in new fundamental phenomena, such as current-induced spin polarization2 and the Spin Hall Effect.3 The electric tunability of SOC has also been achieved by using gated semiconductor heterostructures,4 thereby opening a new gateway to applications ranging from spintronics to quantum computing. Examples of some of the various spintronic devices that have already been studied include spin-field effect transistors,5 spin filters,6 and spin qubit gates.7

Energy-saving spintronic devices are believed to be implementable using persistent spin helix (PSH) materials, as these induce a greatly enhanced spin relaxation time.8–15 Theoretical studies have predicted that such PSH materials can be produced using [001]-oriented quantum wells (QWs) where the Rashba and Dresselhauss terms are equal, or by using [110]-oriented QWs that are only affected by the Dresselhauss Effect.8 In either case, the spin–orbit coupling is linearly dependent on the electron momentum in specific directions, and hence a one-dimensional orientation of the spin textures is generated.8 PSH states have been observed experimentally for [001]-oriented aluminium gallium arsenide (GaAs/AlGaAs) QWs9,10,14 and InAlAs/InGaAs QWs,11–13 as well as for [110]-oriented GaAs/AlGaAs QWs that exhibit uni-directional out-of-plane spin directions.15

Thus far, PSH has only been widely studied in zinc-blende semiconductors. However, experimental observations of high-quality two-dimensional (2D) systems in wurtzite-structured semiconductors16,17 have led to them being increasingly viewed as a promising candidate for spintronics applications.18 As such, there is clearly great value in achieving PSH in these semiconductors. In this letter, we demonstrate that PSH is possible using a wurtzite ZnO(1010) surface. By performing first principles density functional theory (DFT) calculations of the ZnO(1010) surface, a quasi-one-dimensional orientation of the spin textures is identified and clarified by using a simplified spin–orbit Hamiltonian. Finally, it is revealed that the wavelength of this PSH is smaller than those observed in various zinc-blende QW structures.

First principles electronic structure calculations based on the DFT were carried out within a generalized gradient approximation (GGA)19 using OpenMX code.20 In past studies, it was found that the optimized lattice parameters of bulk ZnO are \( a = 3.284 \text{ Å} \) and \( c/a = 1.615 \).18 Surface calculations were carried out using a slab model consisting of 20 bilayers, and were terminated by hydrogen atoms on its backside [Fig. 1(a)]. The vacuum length was set to more than 15 Å in order to avoid interactions between neighboring slabs. The geometries were fully relaxed until the force acting on each atom was less than 1 meV/Å. Norm-conserving pseudo-potentials21 were used. The wave functions were expanded by a linear combination of multiple pseudo-atomic orbitals (LCPAOs) generated using a confinement scheme,22,23 defined as \( \text{Zn6.0-s}^2p^2d^2, \text{O5.0-s}^2p^2d^1, \) and \( \text{H5.0-s}^2p^1 \). For example, in the case of a Zn atom, \( \text{Zn6.0-s}^2p^2d^2 \) means that in this confinement scheme, the cutoff radius is 6.0 bohr,22,23 and two primitive orbitals for the s, p, and d components are used. SOC was included in these fully relativistic calculations, and the spin textures in \( k \)-space were calculated using the \( k \)-space spin density matrix of the spinor wave function.24

Calculations for a ZnO(1010) surface revealed that the out-of-plane relaxation of the Zn and O atoms were \( \delta_z(\text{Zn}) = -0.28 \text{ Å} \) and \( \delta_z(\text{O}) = -0.035 \text{ Å} \), respectively [Fig. 1(a)].
These values were slightly lower than the previous experimental values of $\delta(Zn) = -0.45$ Å and $\delta(O) = -0.05$ Å, but are in a good agreement with past calculations $[-0.36$ to $-0.21$ Å ($\delta(Zn)$) and $-0.04$ Å ($\delta(O)$)]. On the contrary, the in-plane relaxations of the Zn and O atoms, $\delta(Zn) = 0.19$ Å and $\delta(O) = -0.03$ Å, respectively, were close to prior calculations $[\delta(Zn) = 0.116$ Å and $\delta(O) = -0.024$ Å]. The occupied surface states were observed in band structures in the energy range of $-1.3$ to $-0.65$ eV [Figs. 1(b) and 1(c)], from which the partial density of states (PDOS) was calculated through projection onto the surface atoms [Fig. 1(d)]. This revealed that the occupied surface state was characterized by O-2p orbitals, which is consistent with the results of past calculations using local density approximation (LDA).

Given that the surface states are occupied, doping is expected to create a p-type system. Indeed, several researchers have succeeded in producing p-type, non-polar wurtzite ZnO films, lending credence to the notion of creating a p-type wurtzite ZnO(1010) surface.

The effect of SOC on surface states can be seen in Fig. 2, where band splitting is small in the $\Gamma$–$Y$ direction, but quite large in the $\Gamma$–$X$ direction. In these spin-split surface state bands, we find that the spin textures exhibit a quasi-one-dimensional orientation in the in-plane y-direction [Fig. 3(a)], and yet have out-of-plane z components [Fig. 3(b)]. These quasi-one-dimensional spin textures are expected to generate current in a direction perpendicular to the spin orientation and induce a greatly enhanced spin relaxation time through the PSH mechanism. This is supported by the fact that a similar PSH has been observed in [110]-oriented zinc-blende QWs with out-of-plane spin orientations.

To better understand the origin of the spin textures, we must consider the SOC of surface states based on group theory. This states that the ZnO(1010) surface belongs to the symmetry point group $C_{x}$: the mirror reflections in operation in this symmetry transform $(x, y, z)$ to $(-x, y, z)$. Given that a Hamiltonian SOC is completely symmetric in the $C_{x}$ point group and includes first-order terms over the wave vectors, SOC can be expressed as $H_{SOC} = \alpha_{k}k_{x}\sigma_{x} + \alpha_{k}k_{y}\sigma_{y} + \alpha_{k}k_{z}\sigma_{z}$, where $k_{x}$ and $k_{y}$ are wave vectors in the $x$- and $y$-directions, respectively, $\sigma_{x}$, $\sigma_{y}$, and $\sigma_{z}$ are Pauli matrices, and $\alpha_{x}$, $\alpha_{y}$, and $\alpha_{z}$ are coupling constants that define spin–orbit strength. Here, $\alpha_{z}$ is characterized by the in-plane electric field $E_{z}$, whereas $\alpha_{x}$ and $\alpha_{y}$ relate to the out-of-plane electric field $E_{x}$ that is due to the surface effect.

In the case of a bulk system oriented in the [1010] direction, the out-of-plane electric field $E_{z}$ vanishes. Consequently, in the $k_{x}$–$k_{y}$ plane, both $\alpha_{x}$ and $\alpha_{y}$ are zero. This leads to a case where only the first term in the $H_{SOC}$ equation remains, meaning that the bands are spin degenerated in the $\Gamma$–$Y$ direction, and the spin textures are oriented in the out-of-plane $z$-direction. (Calculation results of the bulk system oriented on the [1010] direction are available in the online supplementary data at http://stacks.iop.org/APEX/8/073006/a/)

In surface states, on the contrary, a band split is introduced in the $\Gamma$–$Y$ direction due to the third term in the $H_{SOC}$ equation (Fig. 2). Furthermore, as a result of the second term in this equation, a tilting of the spin textures in the in-plane $y$-direction is induced (Fig. 3). It can therefore be concluded that the above spin–orbit Hamiltonian of the surface state matches well with the calculated results, i.e., the band split in the $\Gamma$–$Y$ direction and the tilt of the spin textures.

Since $H_{SOC}$ is strongly affected by the electric field, as mentioned above, the origin of the spin textures can be further clarified by studying the electric polarization. Given that the spin-split surface state is strongly localized in the first two bilayers [Fig. 4(a)], a strong electric polarization is expected to occur in these bilayers. To clarify this, the layer dependence of the electric polarization was calculated using a point charge model (PCM) for Zn$^{2+}$ and O$^{2-}$ ions in the bilayer in order to calculate the polarization difference: $\Delta P = P(c/a, u) - P(c/a, u_{\text{ideal}})$. Here, $c/a$ and $u$ are the lattice constant ratio and internal parameter for a given optimized structure, respectively, and $u_{\text{ideal}} = 0.375$. As shown in Fig. 4(b), this reveals that the strongest electric polarization was identified near the first bilayer.

Electric polarization in the out-of-plane $\Delta P_{y}$ and in-plane $\Delta P_{x}$ directions was calculated to be $0.0777$ C/m$^{2}$ and $-0.081$ C/m$^{2}$, respectively. These values indicate that the electric field in the out-of-plane $E_{x}$ direction was comparable to that in the in-plane $E_{y}$ direction, which would induce a tilting of the spin orientation [Fig. 4(c)]. However, only the in-plane electric polarization was observed in the case of bulk system ($\Delta P_{y_{\text{bulk}}} = -0.027$ C/m$^{2}$). This led to the generation of the in-plane electric field $E_{x}$, but induced a spin orientation in a fully out-of-plane $z$-direction [Fig. 4(d)]. This confirmed that the spin textures in Fig. 3 were consistent with the PCM results. PSH that induces a greatly enhanced spin relaxation time has recently been extensively studied, with our calcula-
Textures and electric polarizations are calculated using the PCM. Schematic view of the spin plane electric polarization and mirror symmetry, rendering it meV Å),12) (2.0 meV Å)13). This large value of φPSH through achieved using a wurtzite well structures. These results indicate that PSH can be smaller than those observed on various zinc-blende quantum relaxation times has been reported in [110]-oriented zinc-blende QWs,35 suggesting that the ZnO(10\(\bar{1}\)0) surface could provide an efficient spintronics device.

The spin–orbit strength of the PSH (\(\alpha_{\text{PSH}}\)), a variable of interest in spintronics device applications, was calculated using the band dispersion in Fig. 2. This revealed that the value of \(\alpha_{\text{PSH}}\) was quite substantial (34.78 meV Å), and much larger than those observed in the PSH of various zinc-blende n-type QWs of GaAs/AlGaAs [(3.5 to 4.9 meV Å),10] (2.77 meV Å)14] and InAlAs/InGaAs [(1.0 meV Å),12] (2.0 meV Å)13]. This large value of \(\alpha_{\text{PSH}}\) should ensure a small wavelength of PSH (\(\lambda_{\text{PSH}}\), which is important to the miniaturization of spintronics devices. The calculated value \(\lambda_{\text{PSH}}\) (0.19 μm) was in fact one order less than that observed in the direct mapping of PSH (7.3 to 10 \(\mu\)m)10) and the resonant inelastic light-scattering measurement (5.5 \(\mu\)m)14) of GaAs/AlGaAs QWs.

In summary, by studying PSH on a ZnO(10\(\bar{1}\)0) surface through first principles DFT calculations, it has been shown that the spin textures exhibit a quasi-one-dimensional orientation. We also found that the wavelength of this PSH was smaller than those observed on various zinc-blende quantum well structures. These results indicate that PSH can be achieved using a wurtzite (10\(\bar{1}\)0) surface or interface with in-plane electric polarization and mirror symmetry, rendering it suitable for spintronics applications.

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Fig. 4. (a) Expected values of spin projected to the atoms in each bilayer. The calculations are performed for the surface state in Fig. 3(a). The top of surface is represented by \(N = 1\). (b) Calculated data of the in-plane and out-of-plane electric polarizations (\(\Delta P_{\perp}, \Delta P_{\parallel}\)) in each bilayer. The electric polarizations are calculated using the PCM. Schematic view of the spin textures and electric fields for the case of the surface (c) and bulk systems (d).