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RESEARCH ARTICLE

First-principles study of surface states in topological insulators Bi_2Te_3 and Bi_2Se_3 :Film thickness dependence

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Using first-principles method, we investigated the electronic states of Bi_2Te_3 and Bi_2Se_3. We showed that both Bi_2Te_3 and Bi_2Se_3 are insulators with a bulk band gap. In contrast, the surface states of Bi_2Te_3 and Bi_2Se_3 films have a metallic band connecting the conduction and valence bands. The films have an energy gap at the Γ point when the film thickness is less than four quintuple layers (QLs), or about 30 Å. The energy gaps are closed at six QLs and four QLs for Bi_2Te_3 and Bi_2Se_3, respectively. We confirmed the metallicity up to nine QLs. Furthermore, we investigated the spin structures of nine-QL films at the Fermi energy in momentum space. We found that both the Bi_2Te_3 and Bi_2Se_3 films have Rashba-type spin textures; i.e., the surface states have spin-polarization. To investigate the spatial distribution of the spin, we decomposed the expected values of the spin for each atom. The expected values of the spin are localized within the third QL from the surface. Our results of nine-QL films clearly show the boundary between the bulk and surface regions.

Keywords: topological insulators; surface state; Bi_2Te_3; Bi_2Se_3; first-principles calculation

1 Introduction

Topological insulators have recently attracted much attention in solid-state physics [1, 2]. These materials behave as insulators in the bulk states but as metals in the surface states. Because the surface states of topological insulators are protected by time-reversal symmetry, spin-up and spin-down electrons flow in opposite directions at the surface. These properties of topological insulators are expected to be applied in low-power-consumption spintronics devices [3], thermoelectric devices [4, 5], and quantum computing [6].

The bismuth chalcogenides, Bi_2Te_3 and Bi_2Se_3, are known as three-dimensional topological insulators, as revealed experimentally [7, 8]. They have topologically protected metallic surface states, called Dirac cone states, in the bulk insulating gap at the Γ point, $\boldsymbol{k} = (0, 0, 0)$. Although the charge distribution of the surface states of Bi_2Te_3 was analyzed theoretically [9], the boundary between the bulk and surface regions were not clearly shown.

In this study, we investigated the electronic states of Bi_2Te_3 and Bi_2Se_3 using first-principles method based on density functional theory. We focused on the film thickness dependence of the surface states of Bi_2Te_3 and Bi_2Se_3. The energy gap at the Γ point was investigated for various film thicknesses. Furthermore, we revealed the spatial distributions of the spin of Bi_2Te_3 and Bi_2Se_3 in the surface states.

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2 Method

Using the OpenMX code [10], we performed first-principles calculations based on density functional theory within the generalized gradient approximation [11]. The spin-orbit interaction is included in this calculation. We used linear combinations of pseudo-atomic orbitals [12, 13]. The cutoff radius of the radial wave function (the length unit is bohrs) and orbitals are set as follows: 8.0-s3p3d2 for Bi, 7.0-s3p3d1 for Te, and 7.0-s3p3d1 for Se. We used the norm-conserving pseudopotentials [14]. The cutoff energies are 200 and 150 Ry in the calculations for the bulk and the surface, respectively. The k-points are sampled as $8 \times 8 \times 8$ in the bulk calculation and $10 \times 10 \times 1$ in the film calculation. To evaluate the spin texture of surface states, we calculated the k-dependent expected values of the spin by the same method as in the previous study [15].

3 Crystal structure

Both Bi_2Te_3 and Bi_2Se_3 have a rhombohedral unit cell, and the space group is $R\bar{3}m$. When we calculate the films of these crystals, we use a hexagonal unit cell. By transforming the lattice vectors, we can obtain a hexagonal unit cell, and the crystals have a layered structure along the c-axis of the hexagonal unit cell. The unit layer consists of X-Bi-X-Bi-X (X = Te, Se), which is called a quintuple layer (QL). To investigate the electronic states of the surface using the films, the vacuum regions are 20 Å for the slab models in Bi_2Te_3 and Bi_2Se_3. Figure 1 (a) shows the crystal structure of the rhombohedral unit cell with the lattice constant a_r and the angle α_r. Figures 1(b) and 1(c) show side and top views of the hexagonal unit cell with the lattice constants a and c, respectively. Table 1 shows the lattice parameters used in this study. We used the experimental lattice parameters [16].

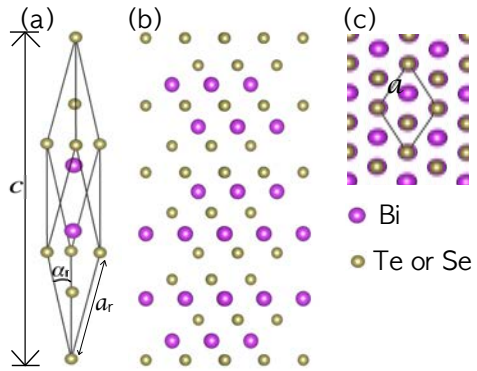

Figure 1. Crystal structure of (a) rhombohedral unit cell, (b) side view and (c) top view of hexagonal unit cell. The lattice parameters are denoted by a_r and α_r for the rhombohedral unit cell and by a and c for the hexagonal one.

Table 1. Lattice parameters of rhombohedral unit cell and hexagonal unit cell [16].

	rhombohedral unit cell		hexagonal unit cell	
	a_r [Å]	α_r [degree]	a [Å]	c [Å]
Bi_2Te_3	10.476	24˚166’	4.386	30.497
Bi_2Se_3	9.840	24˚304’	4.143	28.636

4 Result and discussion

Figure 2 shows the band structures of bulk Bi_2Te_3 and Bi_2Se_3. The high-symmetry points are Γ $(0, 0, 0)$, Z$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, F$(0, \frac{1}{2}, \frac{1}{2})$, and L$(0, \frac{1}{2}, 0)$ for the rhombohedral reciprocal lattice vectors. The

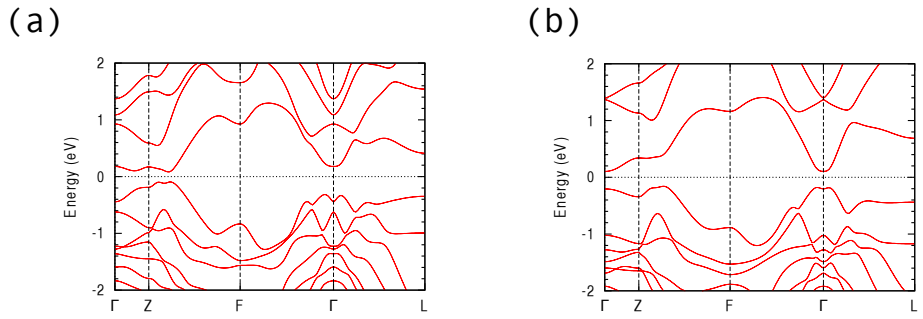

Figure 2. Band structures of (a) Bi_2Te_3 bulk and (b) Bi_2Se_3 bulk.

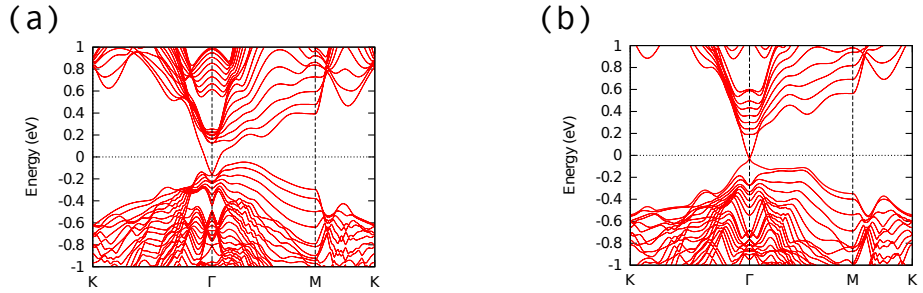

Figure 3. Band structures of nine-QL films; (a) Bi_2Te_3 and (b) Bi_2Se_3.

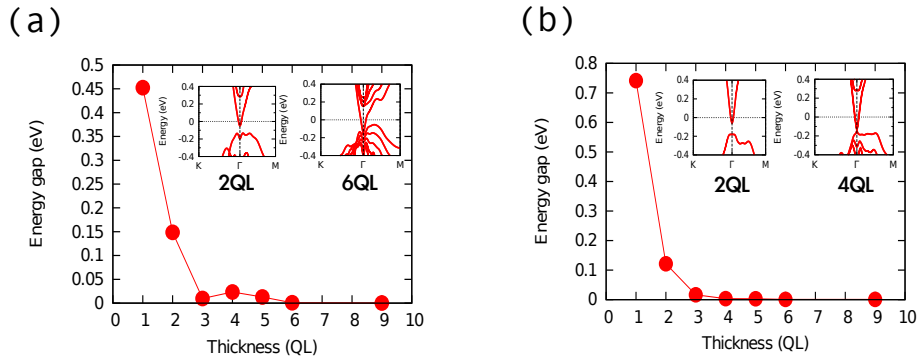

Figure 4. The energy gap at Γ point is plotted as a function of film thickness (QL) for (a) Bi_2Te_3 and (b)Bi_2Se_3; in the respective inset, band structures of typical numbers of QL are shown.

band structures are similar to those obtained in the previous first-principles calculations [17]. In Figure 2, Bi_2Te_3 and Bi_2Se_3 are insulators with energy gaps of 0.17 and 0.26 eV, respectively.

Figure 3 shows the band structures of nine-QL films for Bi_2Te_3 and Bi_2Se_3. The high-symmetry points are Γ $(0,0,0)$, K$(\frac{1}{3},\frac{2}{3},0)$, and M$(0,\frac{1}{2},0)$ for the hexagonal reciprocal lattice vectors. The features of the band structures of Bi_2Te_3 and Bi_2Se_3 are qualitatively similar to those obtained in the previous first-principles calculations for thinner films (five QLs and six QLs) [9, 18]. Bi_2Te_3 and Bi_2Se_3 films have surface states connecting the valence and conduction bands.

Further, we plotted the energy gaps of Bi_2Te_3 and Bi_2Se_3 at the Γ point as a function of the film thickness, as shown in Figure 4. Both Bi_2Te_3 and Bi_2Se_3 have energy gaps when the film thickness is less than four QLs. These gaps are due to the hybridisation between the wave functions of the top and bottom surfaces [19]. On the other hand, the energy gap of Bi_2Te_3 is closed at six QLs, and that of Bi_2Se_3 is closed at four QLs. This film thickness dependence of the band gap is similar to that in a previous study up to six QLs [20, 21].

Next, to investigate the spin structure of the surface states, we plotted the spin textures of nine-QL films at the Fermi energy projected to the first QL from the surface. Figure 5(a) and

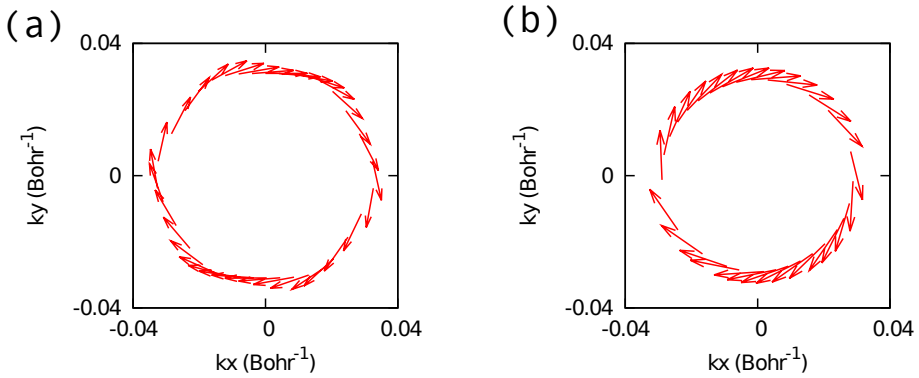

Figure 5. Spin texture of nine-QL films on the plane of the Bloch wave vectors (k_x, k_y) at the Fermi energy for (a) Bi_2Te_3 and (b) Bi_2Se_3. The expected values of spin are calculated using the wavefunctions projected to the atomic orbitals in the first QL from the top surface. The longest arrow corresponds to 0.33 μ_B for Bi_2Te_3, 0.51 μ_B for Bi_2Se_3, respectively.

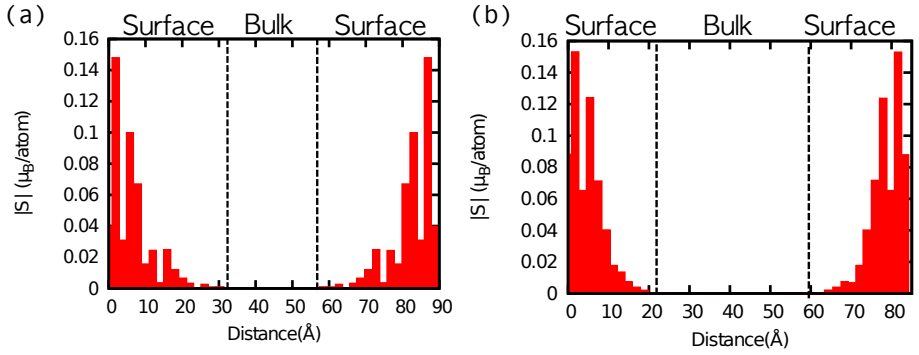

Figure 6. Spatial distribution of spin for (a) Bi_2Te_3 and (b) Bi_2Se_3; The horizontal axis represents the distance from the top surface, and the vertical axis represents expected value of spin |S|.

5(b) show the spin texture of the nine-QL films on the k_x-k_y plane of the Bloch wave vector for Bi_2Te_3 and Bi_2Se_3, respectively. The spins at $\boldsymbol{k}$ and $-\boldsymbol{k}$ are in opposite directions, indicating that the system has time-reversal symmetry. From these results, we confirmed that the Dirac cone states in Bi_2Te_3 and Bi_2Se_3 are protected by time-reversal symmetry; i.e., the materials are topological insulators.

Finally, we investigated the spatial distribution of the spin by projecting the expected value of the spin to each atom. Figure 6(a) and 6(b) show the spatial distribution of the spin for Bi_2Te_3 and Bi_2Se_3, respectively. The films are clearly separated into surface and bulk regions. The surface states are localized within the third QL from the surface. Moreover, the surface region of Bi_2Se_3 is more localized than that of Bi_2Te_3. This is ascribed to the larger band gap in the bulk Bi_2Se_3.

5 Summary

Using the first-principles method, we investigated the electronic states of Bi_2Te_3 and Bi_2Se_3. We confirmed the following properties of these materials. Bi_2Te_3 and Bi_2Se_3 are insulators in the bulk. In contrast, Bi_2Te_3 and Bi_2Se_3 films have surface states connecting the valence and conduction bands. Both Bi_2Te_3 and Bi_2Se_3 have energy gaps when the film thickness is less than four QLs. These results are attributed to hybridisation between the wave functions at the top and bottom of the films. On the other hand, the energy gap of Bi_2Te_3 is closed at six QLs, and that of Bi_2Se_3 is closed at four QLs. The energy gaps of nine-QL films are also closed, and their gapless surface states with spin-polarization in momentum space are localized within the third QL from the surface. Therefore, our results for nine-QL films clearly show the boundary between

the bulk and surface regions.

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