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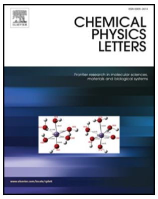

Research paper

# Discovery of a new quantum spin Hall phase in bilayer plumbene

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## ABSTRACT

Two-dimensional (2D) large-gap quantum spin Hall (QSH) insulator are imperative for achieving dissipationless transport devices. The bottleneck preventing applications from QSH phase, however, is lack of feasible 2D films experimentally. By using first-principles calculations, here we propose a new 2D topological phase in the bilayer plumbene. Its geometric stability is confirmed by phonon spectrum simulation and *ab initio* molecular dynamics simulation. We find that it can turn into a nontrivial QSH phase under tensile strain and its nontrivial gap is highly tunable by external strains. The origin behind QSH effect can be identified by $s$–$p_{x,y}$ band inversion, topological invariant $Z_2$, and helical edge states within the bulk gap. Also, a heterostructure composed of bilayer plumbene deposited on semiconducting $MoS_2$ substrate remains topologically nontrivial properties with a sizable gap. These findings provide a promising platform to design a large-gap QSH insulator in Pb film, which show potential applications in spintronics devices.

Two-dimensional (2D) materials have attracted extensive attention in both condense matter physics and material science due to their novel physical properties because the charge and spin are both confined to a 2D plane [1–4]. 2D topological insulators (TIs), also known as quantum spin Hall (QSH) insulators, are more interesting as, even though the bulk material is insulating, the system supports spin-polarized gapless edge states with Dirac-like linear energy dispersion protected by time-reversal symmetry (TRS) [5–7]. Remarkably, the robustness of edge states against the nonmagnetic impurities makes 2D TIs better suited for coherent spin-transport related practical applications in spintronics devices. Since the topological model in graphene is proposed by Kane and Mele [8,9], many ultrathin 2D films, such as Si [10,11], Ge [12], Sn [13–15], As, [16] Bi [17–19], $ZrTe_5/HfTe_5$ [20], and 1*T*'-$MoX_2$ [21], have been put forward theoretically to harbor QSH effect. Unfortunately, the experimental advance of the existence of topological spin transport channels is only limited to HgTe/CdTe [22,23]and InAs/GaSb [24,25] quantum wells at very low temperatures due to weak spin-orbit coupling (SOC). To expand and advance the practical application of 2D films, it is essential to search for new 2D TIs of high working temperatures to overcome the thermal disturbance for future electronic device applications in spintrocs.

Lead (Pb), a group-IV elemental counterpart of carbon, is known for its strong SOC effect, which may drive nontrivial topological states, examples including PbSe monolayer [26], PbC/MnSe heterostrutures, [27] Pb-based chalcogenide series [28] and so on. More recently, topological properties of monolayer plumbene with hexagonal structure have been investigated theoretically [29]. Ma et al. [30] also reported the topological properties in Methyl-functionalized bismuth, antimony, and lead Bilayer Films. Similar to hexagonal configuration in 2D materials [31–33], monolayer plumbene has a buckled honeycomb lattice and shows a metallic state. To produce nontrivially topological states, functionalization has been conducted in monolayer plumbene. Especially, these serial of X-decorated monolayers plumbene have an extraordinarily giant bulk gap of 1.34 eV [29]. Unfortunately, the experimental realizations of the quantum topological states in monolayer case have not reported up to date.

In the present work, on the basis of first-principles calculations, we extend the monolayer plumbene to bilayer one, as shown in Fig. 1(a). The phonon spectrum and *ab initio* molecular dynamics simulations confirm the structure stability of bilayer plumbene, which is possible to synthesis in experiments. Analyses of the electronic structures reveal that it becomes a QSH phase under tensile strain, whose topological electronic properties are highly tunable under external strain. The physics of the QSH effect can be identified by $s$–$p_{x,y}$ band inversion, topological invariant $Z_2$, and helical edge states. Also, we propose a heterostructure by depositing bilayer plumbene on semiconducting $MoS_2$ substrate and find that bilayer plumbene remains topologically nontrivial with a sizable gap. Our results enrich the 2D family of QSH insulator and enable its potential room-temperature applications in spintronic devices.

We perform all calculations by using density functional theory (DFT) [34] as implemented in the Vienna Ab initio simulation package (VASP) [35]. The exchange-correlation interaction was treated within the generalized gradient approximation (GGA) in the form proposed by Perdew-Burke-Ernzerhof (PBE) [36], and the plane-wave basis with a kinetic energy cutoff of 500 eV are employed. The Brillouin zone is sampled by using a 9 × 9 × 1 Gamma-centered Monkhorst-Pack grid. The vacuum space is set to 20 Å to minimize artificial interactions

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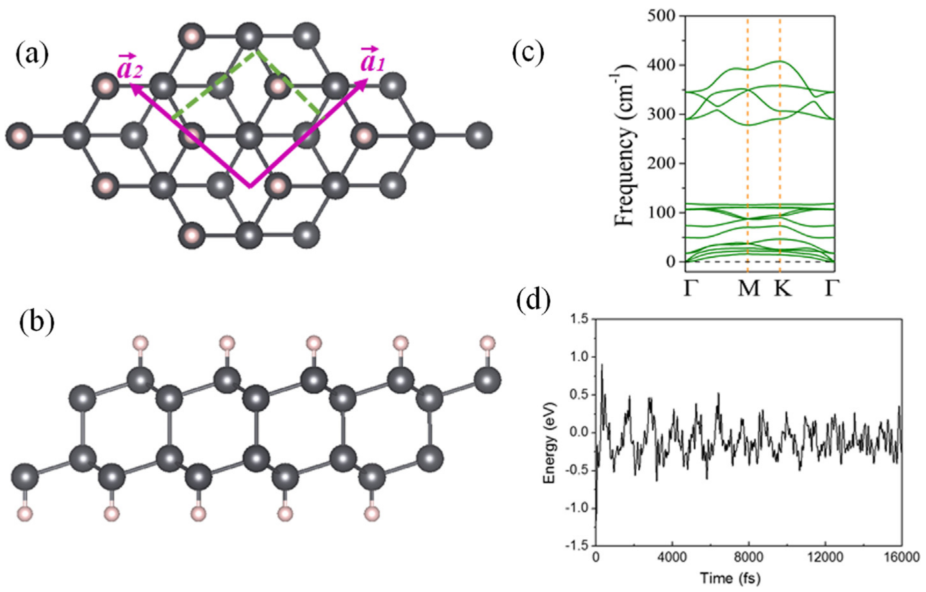

**Fig. 1.** (a) and (b) Top and side view of the bilayer plumbene. Black and cyan balls signify Pb and H atoms, respectively. The shadow area in (a) represents the unit cell. (c) phonon-spectrum of bilayer plumbene, where no soft mode is found. (d) Variations of the energy for bilayer plumbene during MD simulation at temperature of 300 K. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

between neighboring slabs. During the structural optimization, all atomic positions and lattice parameters are fully relaxed, and the maximum force allowed on each atom is less than 0.02 eV/ Å. The screened exchange hybrid density functional by Heyd-Scuseria-Ernzerhof (HSE06) [37,38] is adopted to further correct the electronic structure. The phonon calculations are carried out by using the PHONOPY code [39] through the DFPT approach [40] with SOC.

Fig. 1(a) shows the geometric structure of the bilayer plumbene, in which all nearest neighboring Pb atoms are sp$^3$ hybridized. Interestingly, the optimized buckled height of 0.82 Å is found to be similar to the case (0.78 Å) of monolayer plumbene. To evaluate the stability of bilayer plumbene, we calculate its formation energy written as

$$\Delta E_f = E(\text{bilayer}) - E(\text{Pb}) - 1/2E(\text{H}_2)$$

where $E$(bilayer) and $E$(Pb) are the total energies of bilayer plumbene and Pb atom, respectively, while $E(\text{H}_2)$ is the chemical energy of molecule hydrogen. According to our computations, bilayer plumbene is found to have a cohesive energy of 3.52 eV, larger than that of monolayer plumbene. This reveals that the bilayer plumbene is a strongly bonded network. The dynamic stability of 2D bilayer plumbene has been checked by phonon spectrum calculations. As seen in Fig. 1c, all branches have positive frequencies and no imaginary phonon modes, suggesting that the monolayer structure of plumbene is dynamically stable.

To further check the thermal stability of bilayer plumbene, we perform *ab initio* molecular dynamics (MD) simulation at temperature of 300 K, as shown in Fig. 1(d). Note that the structure of bilayer plumbene does not collapse throughout a 16 ps MD simulation up to 300 K, indicating that the melting point of bilayer plumbene lattice is probably at 300 K. The above results reveal that the bilayer plumbene has very good thermal stability and can maintain its structural integrity in a high-temperature environment.

The electronic band structures calculated for 2D bilayer plumbene with SOC in the Brillouin zone (BZ) are shown in Fig. 2. Note that the band structure has a direct band gap of 0.47 eV at the Γ point. An orbit-projected analysis for the composition of the electronic states reveals that in the vicinity of the Fermi level, the top of valence band arises mainly from $p_{x,y}$ orbitals, while the bottom of the conduction band consists of $s$ orbital, as shown in Fig. 2(a). Thus, no inverted band order is observed in this 2D lattice, showing that the bilayer plumbene is a trivial insulator.

The external strain plays an important role to tuning electronic properties of 2D films. Here we employ an in-plane strain perpendicular to $z$ axis maintaining the crystal symmetry by changing its lattices as $\varepsilon = (a - a_0)/a_0$, where a ($a_0$) is strained (equilibrium) lattice constants. Fig. 3 shows the evolution of band gap as a function of strain for bilayer plumbene. With increasing the strain, we find that the CBM is driven continuously to shift downward to the Fermi level, while the VBM increase reversibly, leading the band gap to decrease significantly. Throughout the changes under tensile strain, it has a direct band gap with the VBM and CBM both located at the Γ point. Notably, at the critical value of 6.0% (see Fig. 2(b)), the $p_{x,y}$ and $s$ bands of Pb atoms touch each other at the Fermi level, which can be considered as a gapless semiconductor, or alternatively, as a semi-metal with zero density of states at the Fermi level. When the strain increases beyond 6.0% (see Fig. 2(c)), the most pronounced feature is that the top of valence band derives from $p_{x,y}$, while the bottom of the conduction band from $s$ orbitals near the $\Gamma$ point, suggestive of the band inversion with a large inverted gap. The evolution of band gap changing from open to close to reopen with respect to the lattice constant suggests a topological transition in bilayer plumbene.

To check whether it show nontrivial topology for bilayer plumbene, we calculate the $Z_2$ invariant $\nu$ based on the method proposed by Fu and Kane [41], written as

$$\delta(K_i) = \prod_{m=1}^{N} \xi_{2m}^{i},\ (-1)^{\nu} = \prod_{i=1}^{4} \delta(K_i) = \delta(\Gamma)\delta(M)^3$$

where δ is the product of parity eigenvalues at the time-reversal-invariant momenta (TRIM) points, $\xi = \pm 1$ are the parity eigenvalues and N is the number of the occupied bands. According to the $Z_2$ classification, $\nu = 1$ characterizes a topologically nontrivial phase and $\nu = 0$ means a topologically trivial phase. Here, the invariants $Z_2$ are derived from the parities of wave function at the four TRIM points $K_i$, including one Γ point and three M points in the Brillouin zone, i.e., at (0, 0), (1/2, 0), (0, 1/2), and (1/2, 1/2) TRIMs. In this respect, the topological invariant $z_2$ of bilayer plumbene is found to be +1 in the $\varepsilon = 6.0\%$, indicating that it becomes a nontrivial topological state.

The existence of topologically protected chiral edge states is one of the most important signatures of the QSH effect. To reveal the nontrivial topological nature of the bilayer plumbene, by constructing the Green's functions for the semi-infinite boundary based on the maximally localized Wannier function method [42], we can obtain the local density of state (LDOS) of the edge states, as shown in Fig. 4(c) and (d). Note that all the edge bands connect completely the conduction and valence bands and span the bulk gap, yielding a 1D gapless edge states. Besides, the counter-propagating edge states exhibit opposite spin