Current Applied Physics 18 (2018) 1393–1398

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

Low-energy band structure very sensitive to the interlayer distance in Bernal-stacked tetralayer graphene

Kyu Won Lee, Cheol Eui Lee*

Department of Physics, Korea University, Seoul, 02841, South Korea

ARTICLE INFO

Keywords:
A. Tetralayer graphene
B. Interlayer couplings
C. Metal-insulator transition
D. Quantum valley Hall effect

ABSTRACT

We have investigated Bernal-stacked tetralayer graphene as a function of interlayer distance and perpendicular electric field by using density functional theory calculations. The low-energy band structure was found to be very sensitive to the interlayer distance, undergoing a metal-insulator transition. It can be attributed to the nearest-layer coupling that is more sensitive to the interlayer distance than are the next-nearest-layer couplings. Under a perpendicular electric field above a critical field, six electric-field-induced Dirac cones with mass gaps predicted in tight-binding models were confirmed, however, our density functional theory calculations demonstrate a phase transition to a quantum valley Hall insulator, contrasting to the tight-binding model prediction of an ordinary insulator.

1. Introduction

In a recent experiment on Bernal-stacked tetralayer graphene (4LG) encapsulated with hexagonal boron nitride, multiple Lifshitz transitions (abrupt topological changes in the connectivity of the Fermi surface) and multiband transport were reported to occur as a function of the charge density and perpendicular electric field, which was well described by a tight-binding model with hopping parameters extracted from the experiment [1]. In another experiment on suspended 4LG, a pronounced insulating state was found at the charge neutrality point, which cannot be described by a tight-binding model predicting a semimetallic band structure, and was attributed to a symmetry-broken state arising from electron-electron interactions [2]. In suspended bilayer graphene, electron-electron interactions have been considered to be responsible for the opening of a band gap with some controversy [3–5]. Even in the absence of a bulk band gap, AB-BA stacking faults were suggested to enable the experimentally-observed insulating states in bilayer graphene [6]. In the experiment on suspended 4LG, it was proved that the AB-BA stacking faults are not consistent with several aspects of the experimental observations [2].

However, we believe that the situation is more subtle in 4LG than in bilayer graphene. In the tight-binding model for Bernal-stacked 4LG, the band structure can be described by two intersecting bilayer graphene-like bands with light and heavy effective masses [7–9]. Hybridization of the two massive bands due to next-nearest-layer hopping opens an energy gap at the intersecting points, leading to a semimetallic band structure where the conduction and valence bands touch each other [1,7,9]. Because very weak next-nearest-layer couplings determine the low-energy band structure, we can conjecture that a small change in the interlayer distance modifies the band structure at low energies, as confirmed in this work.

Under a perpendicular electric field, Bernal-stacked bilayer and trilayer graphenes have been predicted to be a quantum valley Hall insulator with a valley Chern number $C_V = 2$ and 1, respectively [8,10–12]. In the tight-binding model for Bernal-stacked 4LG, a perpendicular electric field was predicted to open a band gap, inducing six Dirac cones but leading to an ordinary insulator with zero valley Hall conductivity [8]. The quantum valley Hall state was predicted to generally occur in asymmetric odd-layer graphenes, which was attributed to an approximate chiral symmetry peculiar to odd-layer graphenes [8]. The quantum valley Hall states in bilayer and trilayer graphenes have been confirmed by density functional theory (DFT) calculations [13,14]. Regarding Bernal-stacked 4LG, short comments only have come to our attention that the maximum band gap can be about 8 meV in DFT studies with local density approximations [15,16].

In this work, we have investigated Bernal-stacked 4LG as a function of the interlayer distance and perpendicular electric field by using density functional theory calculations. By using several types of the exchange-correlation functionals, we will show that the low-energy band structure is very sensitive to the interlayer distance. The semimetallic band structure predicted by the tight-binding models, where the conduction and valence bands touch each other, was achieved only

* Corresponding author.
E-mail address: rscel@korea.ac.kr (C.E. Lee).

https://doi.org/10.1016/j.cap.2018.08.003
Received 11 April 2018; Received in revised form 25 June 2018; Accepted 7 August 2018
Available online 11 August 2018

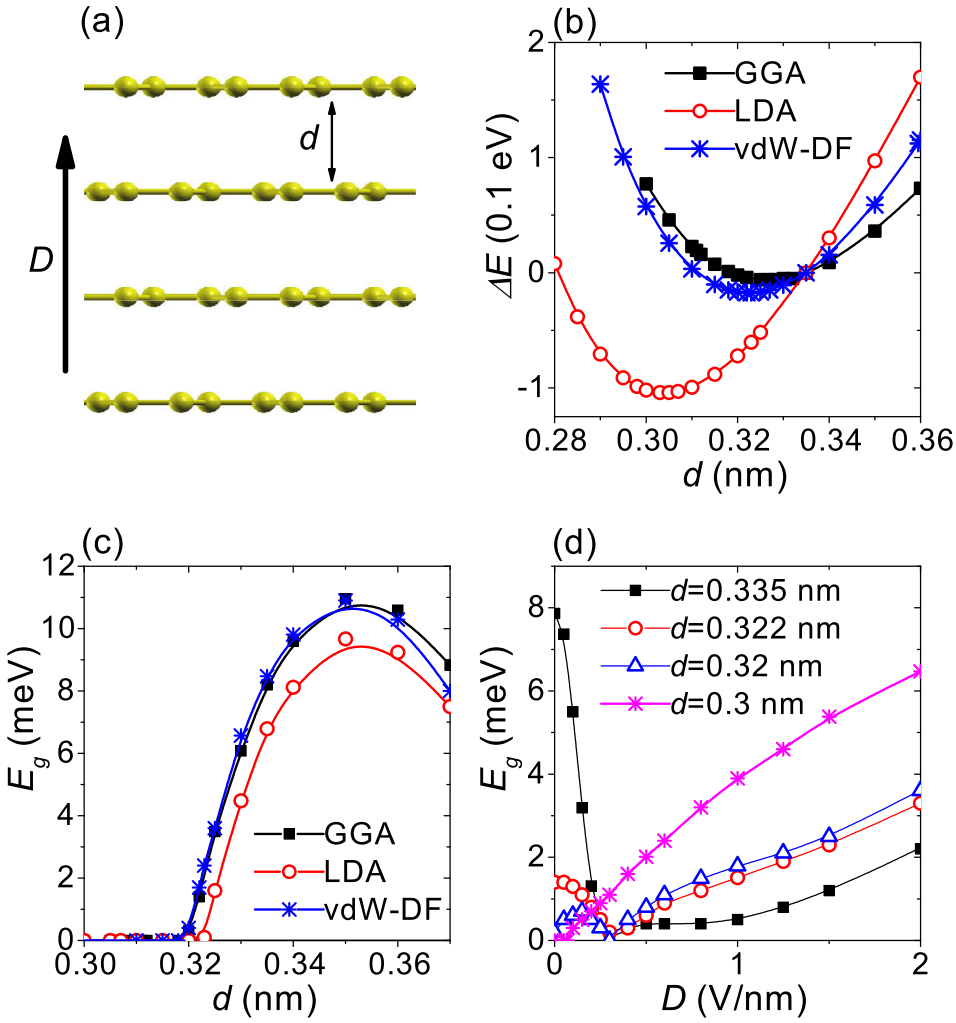

Fig. 1. (a) Schematic structure of Bernal-stacked 4LG. (b) Total energy difference ΔE from the total energy at the interlayer distance $d = 0.335$ nm as a function of d. (c) Band gap E_g as a function of d. (d) E_g as a function of the perpendicular electric field D obtained by using GGA.

at a critical interlayer distance. While six electric-field-induced Dirac cones with mass gaps predicted in the tight-binding models were confirmed, our DFT calculations show that 4LG is a quantum valley Hall insulator with $C_V = 6$ under a perpendicular electric field above a critical field regardless of the interlayer distance, contrasting to the tight-binding model prediction of an ordinary insulator.

2. Methods

Bernal-stacked 4LG was considered as a two-dimensional periodic system with a lattice constant $a = 0.246$ nm. The interlayer distance d was considered as an adjustable parameter. The vacuum spacing between the 4LG was set to 2 nm. A SIESTA package [17], which uses a localized linear combination of numerical atomic-orbital basis sets, was employed for the DFT calculations. A double-ζ polarized (DZP) basis set was used and norm-conserving Troullier-Martins pseudopotentials were used [18]. A local density approximation (LDA) of Perdew and Zunger [19], a generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof [20], and the nonlocal van der Waals density functional (vdW-DF) of Dion et al. as implemented by Román-Pérez and Soler [21–23] were used for the exchange and correlation potential. The plane-wave cutoff energy was set to 350 Ry for the LDA and GGA functionals and 1000 Ry for the vdW-DF. k-points of 100×100×1 mesh in a Monkhorst-Pack scheme were used.

Maximally localized Wannier functions were constructed within the Wannier90 code by using wavefunctions obtained from the DFT calculations [24]. Based on the Wannier interpolation, band structures and Berry curvatures were calculated. The Berry curvature was calculated for the valence bands below the band gap. By using the anomalous Hall conductivity calculation routine implemented in Wannier90 code, the valley Hall conductivity was obtained as the difference between the valley-resolved Hall conductivities, $\sigma_v = \sigma_v(\mathrm{K}) - \sigma_v(\mathrm{K}')$, where $\sigma_v(\mathrm{K})$ and $\sigma_v(\mathrm{K}')$ were calculated by integrating the Berry curvature around the K and K′ points in the first Brillouin zone, respectively. When the valley Hall conductivity $\sigma_v = C_v(e^2/h)$ is quantized, C_v corresponds to the valley Chern number.

3. Results and discussion

Fig. 1(b) shows the total energy difference ΔE from the total energy at the interlayer distance $d = 0.335$ nm as a function of d obtained by using LDA, GGA, and vdW-DF. The total energy has a minimum at $d = 0.305$ nm for LDA, at $d = 0.327$ nm for GGA, and at $d = 0.323$ nm for vdW-DF. Fig. 1(c) shows the band gap E_g as a function of d obtained by using LDA, GGA, and vdW-DF. We can see that the band gap is very sensitive to the interlayer distance, demonstrating a metal-insulator transition at a critical interlayer distance $d_c \simeq 0.32$ nm. While the energetically-stable interlayer distance depends on the exchange-correlation functional used in the DFT calculations, the metal-insulator transition is reproduced regardless of the functionals. Thus, we can confirm that the low-energy band structure of 4LG, which is determined by the weak next-nearest layer couplings, is very sensitive to the interlayer