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Ivan Božović

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A rich new family of quantum materials

Ivan Božović

Shanghai Advanced Research in Physical Sciences (SHARPS), Shanghai 201203, China

Email: bozovic.hts@gmail.com

The discovery of graphene brought to the forefront of condensed matter physics the behavior of electrons near the band crossings where the dispersion is linear, $E(k) = \pm \hbar v_F |k|$; here, E is energy, k is the wavevector measured from the crossing point, $\hbar$ is the Planck constant divided by 2π , and v_F is the Fermi velocity [1,2]. Because of the resemblance to the relativistic Dirac equation, these became known as Dirac dispersion, Dirac crossings, and, in quasi-two-dimensional (2D) materials, Dirac cones. Beyond just a mathematical metaphor, electrons in graphene exhibit very high mobility, the quantum Hall effect at room temperature, and the Klein Paradox tunneling (nearly perfect transmission through high-potential barriers, without backscattering) [3,4].

This motivated the quest for other materials featuring Dirac dispersion, primarily focused on graphene-like honeycomb lattices of other elements (“X-enes”) or compounds (e.g., “MX-enes”, where M is a metal). Other hypothetical hexagonal structures were also explored, including the kagome, Lieb, star, ruby, and Cairo lattice models. A few of these models have found approximate embodiments, but these are still quite rare; as for non-hexagonal Dirac materials, no real-world examples are known [5-10].

The present paper aims to fill this gap. Bulk crystals of alkali, rare-earth, and noble metals often adopt body-centered or face-centered cubic structures. If only two monolayers are deposited on a suitable substrate, these are likely to assume the staggered bilayer square (SBS) lattice arrangement. The SBS lattice has nonsymmorphic symmetry elements and consequently, its band structure entails Dirac crossings. These crossings are of a novel type, giving rise to unipolar anisotropic Dirac conoids and harboring new physics with exotic topological, quantum, and relativistic aspects. This motivates efforts to synthesize SBS lattices of various elements and compounds and study their physical properties.

Nonsymmorphic space-group symmetry elements—screw axes or glide planes with fractional translations along one direction—dictate linear crossings in the

electron band structure [11-14]. Such crossings are symmetry-protected and topologically invariant. They must also be present in the dispersion of other excitations, e.g., in the phonon branches. To better explore this phenomenon, we introduce a staggered bilayer square atomic lattice model (Fig. 1a). The lattice has nonsymmorphic $p4gm$ symmetry, and its band structure features Dirac crossings along specific high-symmetry directions. This model is simple but realistic; it should apply to bilayers of several elemental and compound 2D metals. The physics has some similarities with, e.g., that of the A-B stacked bilayer graphene, but it exhibits novel features, including unipolar, anisotropic Dirac conoids.

Alkali metals (Li, Na, K, Rb, Cs, Fr) feature a half-filled conduction band dominated by s orbitals. Other s^1 metals include Cu, Ag, and Au, as well as Ba (due to a specific hybridization). Most of these assume the body-centered cubic (bcc) structure in the bulk. If only two monolayers are deposited on an appropriate substrate, these could form the SBS structure. In bcc crystals, the M-M bonds are not all equal; to achieve the closest packing (the highest density, maximal cohesion energy), the layers are pushed towards one another at the expense of elongating the in-plane bonds by $2/\sqrt{3} = 1.155$, i.e., about 15%. In a free-standing SBS, this need not happen; the ideal SBS structure should be more stable because it has a higher packing density (0.74, the same as face-centered cubic (fcc) lattice, compared to 0.68 for bcc). In an SBS film on a substrate, some c -axis contraction and a -axis expansion, or other distortions, might happen if the substrate-film interaction is substantial. However, this should not qualitatively change the band structure. Bilayers of s^2 metals, including Be, Mg, Ca, and Sr, could also form the SBS structure on well-chosen substrates.

The SBS model may also be relevant for various compounds. For example, in a one-unit-cell (1 UC) thick layer of NaCl, the sublattice formed by Na^{+1} cations has the SBS structure. It is ‘decorated’ by Cl^{-1} anions, but the first ionization potential of Na is 5.14 eV while that of Cl is 12.97 eV, so the lowest unoccupied band in NaCl is the 3s band of Na, with basically no contribution from Cl orbitals. Thus, the low-energy band structure of 1UC NaCl resembles that of the elemental SBS Na lattice. The same is true for many other ionic compounds in the rock-salt structures and beyond.

1UC thick films of various alkali halides were already synthesized on Ag, Au, Pt, Cu, Ni, and Al substrates. In NaCl, the Na^{+1} ions are relatively far apart, and the Na s -band is very narrow and fully occupied. In this aspect, compounds containing larger alkaline cations and smaller halide anions, such as RbF, are better candidates, since their s -bands are broader. On the other hand, if NaCl is irradiated with electrons, Cl atoms evaporate, leaving behind a Na bilayer in the SBS structure [15]. This points to a new route for synthesizing a variety of SBS films and fabricating nanometer-scale devices, such as metallic nanowires, field-effect transistors, and tunnel junctions.

The generic SBS lattice model is illustrated in Fig. 1a. It consists of two square lattices with one atom per site, staggered halfway along the diagonal. The unit cell contains two atoms, one at $(0, 0, -1/2)$ and the other at $(1/2, 1/2, 1/2)$, in the units $a = b$ and $c = (\sqrt{3}/2)a$. The two are related by the glide plane reflection: $[\sigma_h|_{1/2, 1/2, 0}]$ $(0, 0, -1/2) = (1/2, 1/2, 1/2)$, where σ_h denotes the mirror reflection in the horizontal (x - y) plane. The entire lattice is comprised of two sublattices, bottom and top, with the atoms at $(n, m, -1/2)$ and $(n+1/2, m+1/2, 1/2)$, where $n, m = 0, \pm 1, \pm 2, \dots$. Each atom has eight nearest neighbors, four in the same plane and four in the other plane.

Figure 1. The staggered bilayer square (SBS) lattice model and its electron bands. (a) The SBS lattice is comprised of two sublattices, bottom and top, with the atoms at $(n, m, -1/2)$ and $(n+1/2, m+1/2, 1/2)$, where $n, m = 0, \pm 1, \pm 2, \dots$. The spatial symmetry group is p4gm, and it includes the $[\sigma_h|_{1/2, 1/2, 0}]$ glide plane. Thicker, solid black lines indicate the eight nearest-neighbor bonds. (b) The tight-binding electron band structure of the two-band model, with one s-orbital per atom. The A_0 band (bonding at Γ) and the A_1 band (antibonding at Γ) have a Dirac crossing at X and are degenerate along the X-M direction. (c) A zoom-in on the vicinity of the Dirac crossing at X.

The spatial symmetry group is p4gm, with the generators $[C_4|0,0,0]$, $[\sigma_v|0,0,0]$, $[\sigma_h|_{1/2, 1/2, 0}]$, and the translations $[E|n,m,0]$. Here, C_4 denotes the rotation by $\pi/2$ around the z -axis, σ_v is the mirror reflection in the vertical (x - z) plane, and E is the identity operator. The p4gm group also includes three additional vertical mirror planes (a total of four), four horizontal order-two screw axes, and so on.

The SBS structure can also be viewed as a one-half-unit-cell-thick slice of the fcc lattice, which has the highest density (an occupied volume fraction of 74%). In bulk fcc structures, the coordination number is 12, whereas it is 8 in the SBS lattice. However, this could be compensated for by interaction with the substrate, so we expect that a variety of SBS structures can be realized experimentally.

In the simplest two-band tight-binding approximation, we consider just one s-type atomic orbital $\phi(\mathbf{r})$ per atom. The corresponding crystal orbitals are the Bloch sums:

$$\Psi_k^b(\mathbf{r}) = (1/N) \sum \exp\{i(k_x n a + k_y m a)\} [E|n, m] \phi(\mathbf{r}), \quad (1)$$

$$\Psi_k^t(\mathbf{r}) = (1/N) \sum \exp\{i[k_x (n+1/2)a + k_y (m+1/2)a]\} [\sigma_h|n+1/2, m+1/2] \phi(\mathbf{r}), \quad (2)$$

for the bottom and top planes, respectively, where N is the total number of atoms (we assumed Born-von Karman cyclic boundary conditions, k_x and k_y are the projections of $\mathbf{k}$ onto the x - and y -axis, and a is the lattice constant. For simplicity, we take all the bond lengths to be equal and consider just one hopping integral t between the nearest-neighbor (NN) atoms. For s orbitals, a typical value may be $t_{\text{NN}} \approx 0.5\text{--}1.5$ eV. The p-p hopping integral (t_{pp}) is typically smaller by an order of magnitude. For both s-s and p-p bonds, the next-nearest-neighbor (NNN) hopping is another order of magnitude smaller ($\sim 0.1 t_{\text{NN}}$).

Denoting the on-site energy as E_0 , the NN hopping integral (matrix element) as t , and $\gamma = \exp(ik_x a)$, $\delta = \exp(ik_y a)$, $\eta = \exp(ik_x a/2)$, $\theta = \exp(ik_y a/2)$, we arrive at the following secular equation:

$$\begin{vmatrix} [H_{\text{bb}}(\mathbf{k}) - E] & H_{\text{bt}}(\mathbf{k}) \\ H_{\text{tb}}(\mathbf{k}) & [H_{\text{tt}}(\mathbf{k}) - E] \end{vmatrix} = 0, \quad (3)$$

with the matrix elements:

$$H_{\text{bb}}(\mathbf{k}) = H_{\text{tt}}(\mathbf{k}) = E_0 + t(\gamma + \gamma^* + \delta + \delta^*) = E_0 + 2t[\cos(k_x a) + \cos(k_y a)], \quad (4)$$

$$H_{\text{bt}}(\mathbf{k}) = H_{\text{tb}}(\mathbf{k}) = t(\eta\theta + \eta^*\theta^* + \eta\theta^* + \eta^*\theta^*) = 4t[\cos(k_x a/2)\cos(k_y a/2)]. \quad (5)$$

Eq. (3) has two solutions:

$$E_{\pm}(\mathbf{k}) = E_0 + 2t[\cos(k_x a) + \cos(k_y a) \pm 2\cos(k_x a/2)\cos(k_y a/2)]. \quad (6)$$

The corresponding band structure is sketched in Fig. 1b, for the representative values $E_0 = -10$ eV and $t = -0.5$ eV, along the high-symmetry directions. Since we have two atoms in the unit cell and one atomic orbital per atom, there are two bands. The A_0 band is bonding at the Γ point, even with respect to $[\sigma_h | \frac{1}{2}, \frac{1}{2}, 0]$, and has the bandwidth $W_B = 12t$. The A_1 band is antibonding at the Γ point, narrower ($W_{AB} = 4t$), and odd with respect to $[\sigma_h | \frac{1}{2}, \frac{1}{2}, 0]$, so it has a nodal plane. The latter is topologically invariant; if we continuously tune the model parameters (E_0 and t), the width and shape of the A_1 -band change, but the nodal plane is retained.

The dispersion along ΓM is larger than along ΓX , and larger for the A_0 -band than for the A_1 -band. The two bands are degenerate at both the Γ and X (Y) points. The dispersion from X towards Γ is linear and the slopes of the A_0 - and A_1 -bands are equal in magnitude and opposite in sign, so there is a linear (Dirac) crossing at X , as highlighted in Fig. 1c. This is a straightforward consequence of band back-

folding dictated by the fractional translation included in the non-symmorphic symmetry elements such as $[\sigma_h | \frac{1}{2}, \frac{1}{2}, 0]$ or $[U | \frac{1}{2}, \frac{1}{2}, 0]$, where U is order-two rotation around the x -axis [11]. The upper crossing at M is a topologically equivalent but highly deformed variant of the same phenomenon. The slope at M is zero for both bands, but that is accidental. The degeneracy between the A_1 -band edges at X and Γ is also accidental, meaning that it can be lifted, without breaking the $p4gm$ symmetry, by adjusting the parameters, e.g., by including the NNN hopping. However, the degeneracy of the A_0 - and A_1 -bands along the entire XM (and YM) line is dictated by symmetry. This can be proven by using group-theoretical techniques (i.e., by analyzing the unitary irreducible representation of the $p4gm$ group), but we can offer a simpler argument. In Eq. (6), when $k_x = \pi/a$, $\cos(k_x a/2) = \cos(\pi/2) = 0$, so $H_{bt}(\pi/2a, k_y) = 0$ and hence $E_+(\pi/2a, k_y) = E_-(\pi/2a, k_y)$ for every k . It is straightforward to verify that the same is true for the second-nearest neighbors, and so on, since this indeed follows from the glide-plane and translation symmetry.

In Fig. 2a, we show the shapes of the Fermi surfaces (FS) of the s -orbital bands in a slightly hole-doped SBS lattice. The Fermi level E_F is near the top of the A_0 - and A_1 -bands. Both FSs are centered near the M point, C_4 -symmetric, and hole-like. The FS of the A_0 -band is smaller and almost rectangular; the one of the A_1 -band is larger and resembles a four-leaf clover. For $t = -0.5$ eV and $a = 3$ Å, the effective masses near the top are $m^* = -0.51m_e$ for the A_0 -band and $m^* = -1.53m_e$ for the A_1 -band. The Fermi velocities are small and vanish as $\mathbf{k}$ approaches the M point.

Figure 2. Electronic spectrum of the SBS model. (a) Fermi surfaces for the A_0 and A_1 bands in the case when E_F is slightly below the top of the band (hole doping). The dispersion is parabolic and downward (hole-like) for both bands. The A_0 -band states are even with respect to the horizontal glide-plane reflection, while the A_1 -states are odd and orthogonal to the A_0 -states. (b) Fermi surface for the case when E_F is slightly below the Dirac crossing point at X . (c) Fermi surfaces for E_F slightly above the crossing. (d) Above the crossing, the bands form a unipolar linear-parabolic Dirac conoid. Here, k_x and k_y are the wavevectors measured from the X point. (e) Near the crossing at X , the lower-band dispersion is saddle-like.

In Fig. 2b, we assume that E_F is slightly below the band crossing at X . There is only one large FS, from the A_0 -band. It is centered around Γ , but hole-like since $m^* = \hbar^2/(\partial^2 E/\partial k^2)_{E_F}$ is negative. This band is broad, so these holes should behave as a standard Fermi Liquid. Taking as an example $t = -0.5$ eV, $a = 3$ Å, we get $v_F = 9.6 \times 10^5$ m/s and $m^* = -1.5 m_e$, where $m_e = 9.1 \times 10^{-31}$ kg is the free electron mass.

However, as soon as E_F is slightly above the Dirac crossing at X , the situation changes dramatically. The FS of the A_0 -band expands, traverses the van Hove singularity at X , and becomes open. Two new FSs appear from the A_1 band, both electron-like: a small circular one centered at Γ and an anisotropic one centered at

X, as shown in Fig. 2c. Along Γ -X and near X, the Fermi velocities are $v_F = \pm 4.8 \times 10^5$ m/s, for the A_0 (+) and A_1 (−) bands, respectively.

Along the k_x (Γ -X) direction, and near the X point, the A_0 - and A_1 -bands form a Dirac cross (Fig. 2d), like in graphene. Just above the crossing, the upper Dirac band shape resembles a Dirac cone. Unlike in graphene, it is quite anisotropic — it is V-shaped when viewed along X-M, but parabolic when viewed along the Γ -X direction. This shape may be parametrized as $z = |x| + y^2$ and called ‘linear-parabolic conoid’. Unusually, this Dirac conoid is unipolar — it only extends upwards (electrons only; no holes). This is a new variety of Dirac conoid, neither of the types I, II, or III that were described so far [5,6,8]. The effective Hamiltonian, near the X-point, can be cast as:

$$\hat{H}_{\text{eff}} = (E_0 - tk_y^2)\sigma_0 - 2t(k_x - \pi/a)\sigma_1, \quad (7)$$

where σ_0 is the identity matrix, and σ_1 is the Pauli matrix (and analogously near the Y point).

In the X- Γ direction, the A_0 -band also disperses linearly, just downward, so in this sense, these are also Dirac fermions (holes). However, it disperses parabolically upward in the X-M direction. Rather than a conoid, this surface, $z = -|x| + y^2$, is saddle-like (Fig. 2e). It looks like a pair of folded and deformed planes, and it is cusped where they meet. The A_0 - and A_1 -band touch along the X-M direction, but do not cross. Finally, near the third FS, electron-like and centered at Γ , the A_1 -band disperses upward parabolically, with $m^* = 0.75m_e$ in the Γ -M direction and $m^* = 1.5m_e$ in the Γ -X (or Γ -Y) direction.

Altogether, at this band filling, we should have a three-component electron system, with ordinary mobile holes from the large A_0 -band FS coexisting with the (also ordinary) electrons in the small Γ pocket and with nonstandard, highly mobile Dirac-like electrons from the pockets centered near X (or Y) in topologically nontrivial states.

A comment about the p-bands: The p-band manifold should extend by $8t_{\text{pp}}$ below a higher atomic level, E_p . If the s- and p-manifolds do not overlap, undoped free-standing SBS would be an insulator, with a gap of $\Delta = E_p + 8t_{\text{pp}} - E_s - 4t_{\text{ss}}$. However, if the two manifolds overlap, the physics would then be quite different. The material would be a semimetal, with holes near the top of the two s-bands close to the M point, and electrons near the bottom of the p-bands close to the Γ point. On the other hand, if in a certain SBS material, E_F cuts the two bands derived from p_z -orbitals, separated from the other bands, then the symmetry, band structure, and physics would be similar to what we have described here.

Using a very simple SBS model, we have illustrated how rich new physics, featuring exotic topological, quantum, and (pseudo-)relativistic attributes, may lie within reach in ultrathin films of elemental metals on appropriately chosen substrates. In particular, SBS structures should host Dirac crossings but with unusual dispersion, forming unipolar anisotropic Dirac conoids, and hence the transport and other electronic properties should be anisotropic.

Another new aspect is that in an SBS lattice near half filling, Dirac electrons coexist and exchange with ordinary electrons (Fermi-Liquid quasiparticles from the wide A_0 bonding band). The coexistence of narrow electron bands and local magnetic moments with a sea of fast electrons from a broad band has been explored fruitfully in Kondo Lattice and Periodic Anderson models applied to Heavy-Fermion materials, topological Kondo insulators, valence-fluctuating compounds, and some unconventional superconductors. Coexistence of Dirac and normal band electrons has also been explored in multiband Dirac systems and in the context of hybridization between topological Dirac surface states and normal, parabolic bulk electrons. The situation here is quite different. The parity with respect to the $[\sigma_h|^{1/2}, 1/2, 0]$ glide reflection is a good quantum number and must be conserved [11]. Dirac and normal electrons cohabit in space and exchange. Still, the electron states giving rise to the A_1 band are odd while those of the A_0 band are even with respect to the glide-plane reflection, so they are orthogonal to one another and do not hybridize. Interband scattering is suppressed, leading to enhanced (Klein) tunneling. This can be cast in a pseudospin language, as was done for graphene, using the $SU(2)$ pseudospin symmetry group (expanding to $SU(4)$ if the electron spin is included). If this symmetry is broken externally, e.g., by the interaction with the substrate, this can be described by introducing a pseudo-vector potential $\mathbf{A}_{ps}$ and the corresponding pseudo-magnetic field $\mathbf{B}_{ps}$. One nontrivial physical consequence may be the Quantum Hall Effect in the absence of any real magnetic fields. Other electronic properties, such as electrical conductivity, classical and quantum Hall effect, magnetoresistance, quantum oscillations, thermopower, plasmon spectra and dispersion, optical conductivity, etc., are all expected to be quite unusual.

Intriguingly, several of the elemental metals on our list become superconducting under high pressure; e.g., in Ca, T_c reaches 29 K. It may be possible to reproduce this at ambient pressure by epitaxial or chemical strain. Reduced dimensionality may favor enhanced electron pairing and superconductivity. In $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4/\text{La}_2\text{CuO}_4$ bilayers, with neither material superconducting *per se*, interface superconductivity appears with T_c substantially (25%) higher than in any single-phase $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ sample, at any doping level x . This motivates the study of various metallene/substrate combinations with different charge transfer, physical properties, and excitations in the substrate next to the interface.

Finally, unlike its natural version, twisted-bilayer graphene at certain magic angles becomes superconducting due to the moiré pattern of sparse contacts between

the two layers, the formation of a flat band, and strongly enhanced electron correlations. In our case, moiré patterns can emerge without any exfoliation, repositioning, or twisting, just due to the incommensurability of the metallene lattice with the substrate surface.

Hoping to reinvigorate the interest in 2D quantum materials, we point to the yet unexplored space of new Dirac materials and phenomena, and indicate several research directions in which this work could be extended: (1) The simple model presented here can be generalized, e.g., by varying the vertical distance between the two planes, making $t_{bb} = t_{tt} \neq t_{bt} = t_{tb}$. Suppose the in-plane lattice constant was elongated compared to the shortest distance between the atoms (the M-M bond length), as is the case in the bcc metals, then $|t_{bb}| < |t_{bt}|$. Consequently, at Γ -point, the A_0 -band would move down, and the A_1 -band would move up. The small FS around the Γ point may disappear; however, the crossings at X and M would not shift. (2) If we include the p-orbitals, the key consideration is whether the s- and p-manifolds would overlap. If they do, the SBS slab would be a semimetal, with an indirect s-p band gap. (3) *Ab initio* density functional theory (DFT) calculations, structural optimization, and stability checks can be used to systematically and quantitatively explore SBS lattices of various elements and compounds. (4) Metallic substrates, including Au, Ru, Mo (more inert), or Ag, Cu, and Al (more reactive), have been used to deposit graphene, borophene, and other X-enes. For s^1 and s^2 metals, one should beware of forming alloys or intermetallic compounds. In this respect, Mo may be the best and could support Li, Na, K, Rb, Mg, Ca, Sr, Ba, Au, Ag, Cu, and Al SBS slabs, without alloying. Ru is the next best and can serve for Na, K, Rb, Au, Ag, and Cu. In addition, Pt, Cu, or Al could support Na, K, and Rb.

In addition to chemical stability, one must consider epitaxial strain, structural modifications, and superstructure formation. For most of the above chemically compatible pairs, there is a significant lattice mismatch so that one can expect superstructure modulation, unit-cell enlargement, rotational misalignment, domain formation, moiré physics, and flat-band formation. The substrate surface need not be a square lattice; indeed, micrometer-size single-crystal domains of borophene were successfully grown on both hexagonal Cu(111) and tetragonal Cu(100) surfaces. DFT can be used to elucidate the metallene/substrate interaction, structural reconstruction, and electron transfer across the interface.

The numerous new Dirac materials proposed here should span a wide range of chemical, mechanical, thermal, electronic transport, magnetic, and optical properties. They could be used as building blocks in heterostructures, e.g., by placing exfoliated graphene, X-enes, or MX-enes on top. That could greatly expand the list of possible heterostructures, offering great flexibility to tune the device functionalities and applications.

Experimentally, we can synthesize SMS metallenes by physical deposition, molecular beam epitaxy (MBE), chemical vapor deposition (CVD), atomic layer deposition (ALD), or the submersion/resurfacing method. Then we need to determine the atomic structure, e.g., using *in situ* tools such as reflection high-energy electron diffraction (RHEED) or low-energy electron diffraction (LEED). *Ex situ*, one could use cryogenic atomic force microscopy (AFM), scanning tunneling microscopy (STM), angle-resolved photoemission spectroscopy (ARPES), X-ray diffraction (XRD), transmission electron microscopy (TEM), etc. The metallenes are reactive (except perhaps for SBS goldene), but this may be managed by using a vacuum suitcase or by *in situ* encapsulation (e.g., with Se), transferring the sample to the characterization chamber, and heating it gently to evaporate the cap layer (Se). Next, SBS metallene films should be deposited on semiconducting or insulating substrates such as Si or sapphire, or lifted off metallic substrates and transferred onto insulating ones. This would enable transport, Hall effect, and magneto-transport measurements, fabrication of field-effect (FET) and electrolyte-double-layer (EDLT) transistors, etc.

Conflict of interest

The author declares that he has no conflict of interest.

Acknowledgments

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