Van der Waals metamaterials

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(Dated: October 1, 2019)

Van der Waals heterostructures are a fertile frontier for discovering emergent phenomena in condensed matter systems. They are constructed by stacking elements of a large library of two-dimensional materials, which couple together through van der Waals interactions. However, the number of possible combinations within this library is staggering, and fully exploring their potential is a daunting task. Here we introduce van der Waals metamaterials to rapidly prototype and screen their quantum counterparts. These layered metamaterials are designed to reshape the flow of ultrasound to mimic electron motion. In particular, we show how to construct analogues of all stacking configurations of bilayer and trilayer graphene through the use of interlayer membranes that emulate van der Waals interactions. By changing the membrane's density and thickness, we reach coupling regimes far beyond that of conventional graphene. We anticipate that van der Waals metamaterials will explore, extend, and inform future electronic devices. Equally, they allow the transfer of useful electronic behavior to acoustic systems, such as flat bands in magic-angle twisted bilayer graphene, which may aid the development of super-resolution ultrasound imagers.

I. INTRODUCTION

1 The recent excitement surrounding van der Waals (vdW) heterostructures stems from their ability to display diverse emergent phenomena simply by layering two-dimensional materials like graphene, transition metal dichalcogenides, or phosphorene[1–3]. They are already poised to contribute to transformative technology including ultra-thin low-energy transistors[4], photodetectors[5] and light-emitting diodes[6]. Recently, their capabilities were expanded to include exotic many-body quantum behaviors such as unconventional superconductivity, which occurs in magic-angle twisted bilayer graphene[7,8]. This system exemplifies one advantage of vdW heterostructures: they generally possess simple tuning parameters, such as electrostatic doping, which can alter carrier concentration without introducing complex chemical disorder. However, unearthing such novel systems is an intimidating and time-consuming task, given the countless stacking combinations afforded by the ever-increasing library of two-dimensional materials. An open challenge is to develop a method for rapidly prototyping vdW heterostructures to create a tight feedback loop for their technological advancement.

2 In the last few years, phononic metamaterials have emerged as a promising platform for mimicking simple condensed-matter systems[9–12]. They are appealing quantum mimics because their governing wave equations are straightforward, making calculations fast; they can be quickly fabricated; and their properties derive from their freely-designed structure. Consequently, a carefully constructed phononic metamaterial can host propagating sound waves that closely resemble the behavior of electrons moving in solids. A prominent example of this correspondence is the presence of Dirac-like modes in phononic crystals[13,14], similar to those in graphene (Fig. 1(a-b)). In graphene, these modes rely on the C_6 symmetry of the honeycomb lattice. But this symmetry can be easily reproduced in a phononic metamaterial, for example, by using a honeycomb arrangement of steel pillars (Fig. 1(c)). The resulting phonon band structure of the metamaterial contains a hallmark Dirac cone at the K point (Fig. 1(d)), permitting its use as an accessible probe of Dirac physics. This general framework has been applied to yield analogues of graphene in photonics[15], surface acoustics[16], and mechanics[12,17]. Given their success, it is natural to ask whether the same can be done for multi-layer systems. To date, controllably coupling metamaterials in the manner required to mimic vdW heterostructures remains challenging, despite several promising layered designs[18–20]. Meanwhile, coupling-induced hybridization effects were investigated in systems of neighboring acoustic resonators[21–23], but whether they can be used to imitate vdW heterostructures is an open question.

3 In this paper, we develop a framework for mimicking layered vdW heterostructures by using coupled acoustic metamaterials. Specifically, we numerically explore the coupling strength of an inter-layer membrane as a function of material properties using the commercial finite-element modeling software COMSOL MULTIPHYSICS. We discover recipes for accurately recreating all stacking configurations of bilayer and trilayer graphene. By tuning the membrane properties, we are able to reach regimes far beyond those occurring in graphene, leading to a simple phenomenological model for coupling in our acoustic metamaterial. Our work opens a new path to simulate novel vdW heterostructures, which could uncover new phenomena and inform the fabrication of future vdW materials. In the opposite direction, our work guides the translation of novel electronic vdW phenomena to acoustic systems, which stimulate innovative phononic devices.

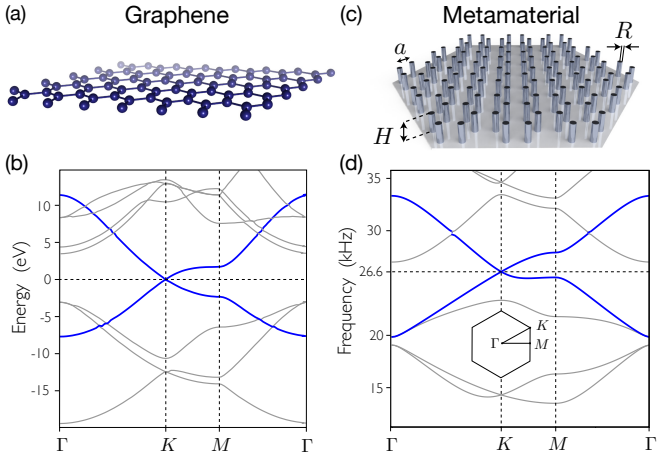

Fig. 1. A phononic metamaterial analogue of graphene. (**a**) In graphene, the honeycomb lattice symmetry leads to (**b**) a band structure featuring a linearly dispersive K-point Dirac cone (blue), adapted with permission from Ref. [24]. (**c**) We designed a phononic metamaterial to imitate graphene by arranging honeycomb lattice of steel pillars in air. The pillars have a radius (R) of 0.32 cm, a height (H) of 0.1 cm, and are spaced (a) by 1 cm. (**d**) Their simulated phononic band structure recreates the Dirac cone in graphene (blue).

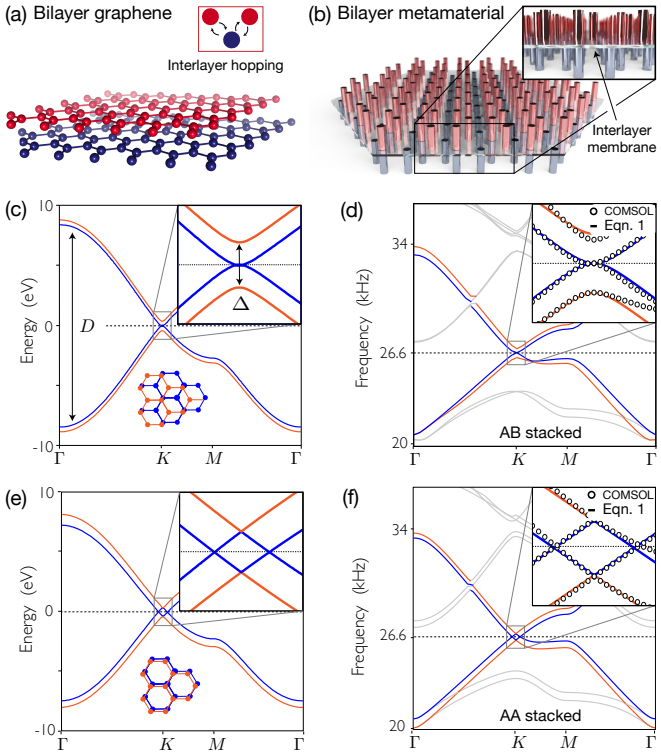

Fig. 2. A phononic metamaterial analogue of bilayer graphene. (**a**) We recreated interlayer hopping in bilayer graphene using (**b**) two layers of honeycomb metamaterial separated by a flexible membrane. (**c**) In AB-stacked bilayer graphene, the Dirac cones hybridize to create a 'kissing' band structure. (**d**) With an interlayer made of 0.22 mm thick HDPE, the same effect is seen in our bilayer metamaterial. This effect is well described by a the same tight-binding Hamiltonian used to describe bilayer graphene (inset). (**e**) In AA-stacked bilayer graphene, the structure of the Dirac cone changes slightly. (**f**) By layering our metamaterial in the same way, it accurately captures this new Dirac cone structure. Panels (c) and (e) are reprinted with minimal alterations from Ref. [25], with permission from Elsevier.

II. PHONONIC BILAYER GRAPHENE

4 Building on previous work, we started from a monolayer phononic metamaterial designed to accurately emulate the Dirac cone in graphene[11,26,27]. The Dirac cone relies only on the C_6 symmetry of the unit cell, but it can be tweaked by adjusting specific material choices. For convenience, our device consists of a honeycomb lattice of steel pillars in air, as shown in Fig. 1(c). We calculated its band structure for various configurations of pillar radius (R), height (H), and spacing (a), and found the closest match to graphene when $R = 0.32$ cm, $H = 0.1$ cm, and $a = 1$ cm (Fig. 1(d)). We focused on mimicking only the graphene Dirac cone, and ignored small differences in the bands around it. Specifically, for frequencies close to 26.6 kHz our acoustic device responds similarly to undoped graphene.

5 Our primary advance is to demonstrate an acoustic analogue of bilayer graphene by stacking two honeycomb metamaterials on top of each other. In its natural state, bilayer graphene arranges in an AB stacking configuration (Fig. 2(a)). Each layer contributes an identical Dirac cone, $E(\mathbf{k})$, to the band structure. The two Dirac cones then become coupled together through interlayer hopping, Δ, to generate a unique band structure whereby the linear Dirac cones are replaced by parabolic 'kissing' bands[25] (Fig. 2(c)). The combined structure can be described by a simple tight-binding Hamiltonian[25,28],

$$\mathcal{H}(\mathbf{k}) = \begin{bmatrix} E(\mathbf{k}) & \delta \\ \delta^T & E(\mathbf{k}) \end{bmatrix}, \quad \text{where} \tag{1}$$

$$E(\mathbf{k}) = \begin{bmatrix} 0 & v_F\mathbf{k} \\ v_F\mathbf{k} & 0 \end{bmatrix}, \quad \delta = \delta_{AB} = \frac{1}{2}\begin{bmatrix} 0 & 0 \\ \Delta & 0 \end{bmatrix},$$

and v_F is the Fermi velocity. In this model, $E(\mathbf{k})$ describes hopping between sublattice sites within a single layer, while δ_{AB} describes the first-order interlayer coupling, which always occurs between inequivalent sublattice sites and so contributes an off-diagonal term. To compare the magnitude of the interlayer hopping to that in our metamaterial, we used a dimensionless coupling metric, $\widetilde{\Delta} = \Delta/D$, where D is the Dirac-cone bandwidth. In bilayer graphene, this ratio was calculated to be about

5%[24].

6 Our metamaterial analogue of bilayer graphene controllably couples phonons in each layer through the use of an intermediary membrane (Fig. 2(b)). Intuitively, as phonons in one layer propagate, they induce matching oscillations in the membrane, which links to the other layer and promotes the desired layer-to-layer crosstalk. To achieve a similar coupling magnitude as bilayer graphene, we used a 0.22-mm thick sheet of high-density polyethylene (HDPE) as the intermediary layer. We computed the phononic band structure of our bilayer metamaterial using COMSOL MULTIPHYSICS (open circles in Fig. 2(d)). Strikingly, the band structure contains 'kissing' bands, analogous to those in bilayer graphene. Furthermore, it is well described by our tight-binding model (Eqn. 1, solid lines in Fig. 2(d)) with reasonable values for $v_F = 140$ m/s and $\Delta = 700$ Hz (or $\widetilde{\Delta} = 5.2\%$), demonstrating that our HDPE sheet accurately recreates the interlayer hopping in bilayer graphene.

7 Here we can demonstrate the first key advantage of vdW metamaterials: the versatility of their stacking configuration. In bilayer graphene, stacking arrangements other than AB are technically challenging to fabricate, yet they host a rich range of physical phenomena[7,29]. On the other hand, exploring translated or twisted metamaterials presents no additional difficulty, allowing analogues of these phenomena to be rapidly explored. To illustrate this point, we recreated the band structure of AA-stacked bilayer graphene by translating our AB-stacked metamaterial, but using the same HDPE interlayer (Fig. 2(e-f)). The band structure is well described by the tight-binding Hamiltonian in Eqn. 1, but with a modified coupling matrix that instead encodes interlayer hopping between vertically-aligned, identical sub-lattice sites:

$$\delta = \delta_{AA} = \frac{1}{2}\begin{bmatrix}\Delta & 0\\ 0 & \Delta\end{bmatrix}. \tag{2}$$

This model accurately describes both the acoustic and electronic AA-stacked systems, as shown in the insets to Fig. 2(e-f). In addition, it captures the behavior of both AA-stacked and AB-stacked metamaterials using the same values of v_F and Δ, simply switching the δ matrix. Our demonstrated success of vdW metamaterials to mimic different stacking configurations of graphene promotes them as an intriguing platform to explore the emergent flat bands in twisted bilayer systems.

8 The second major advantage of vdW metamaterials is the ease with which membrane properties can be changed to explore diverse coupling regimes. In electronic vdW heterostructures, the interlayer coupling is often seen as a fixed property, and experiments that vary it are technically challenging. For instance, it requires substantial longitudinal pressure of 2.5 GPa for approximately a two-fold increase in the coupling strength by pushing hexagonal boron nitride and graphene closer together[31]. Yet, this additional tuning knob has permitted some remarkable discoveries; for example, it can increase the superconducting transition temperature in twisted bilayer graphene by adjusting the magic angle[32]. In our system, tuning the equivalent coupling parameters is as simple as using a thicker membrane or changing its composition. Intuitively, the same pressure variation causes a flexible membrane to move more than a stiff membrane—like how a balloon expands more easily than a basketball when inflated. Consequently, the interlayer coupling increases as the membrane becomes thinner or less dense. We computed the band structure of an AB-stacked bilayer metamaterial to quantify $\widetilde{\Delta}$ as a function of thickness for five different interlayer materials, as shown in Fig. 3. We found that $\widetilde{\Delta}$ does not vary strongly with the speed of sound in the interlayer, but appears to be power-law dependent on its density (ρ) and thickness (T), provided the speed of sound in air is larger than it is in the membrane. It follows the phenomenological rule:

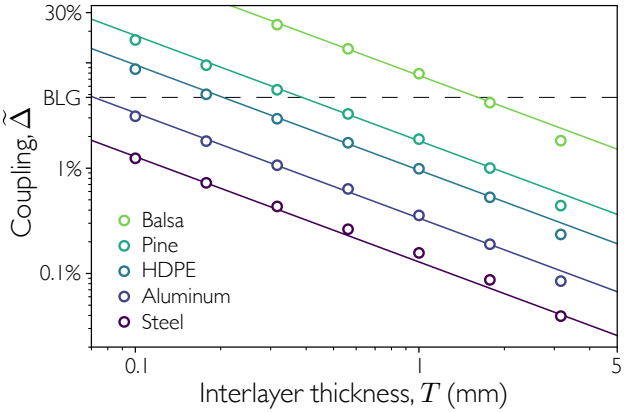

Fig. 3. Interlayer coupling is controllable over two orders of magnitude. The computed dimensionless coupling strength ($\widetilde{\Delta}$, open circles) appears to follow a inverse power law as a function of interlayer density (ρ) and thickness (T): $\widetilde{\Delta} = \Delta/D \propto 1/(\rho T)$ (solid lines), where Δ is the interlayer hopping and D is the bandwidth. With common materials, it is possible to engineer interlayer coupling to be an order of magnitude larger or smaller than in natural bilayer graphene (4.7%, dashed line).

$$\widetilde{\Delta} \equiv \frac{\Delta}{D} \propto \frac{1}{\rho T}. \tag{3}$$

These strong dependencies make it possible to vary $\widetilde{\Delta}$ over more than two orders of magnitude using only common household materials, like steel or wood. Consequently, phononic metamaterials present themselves as feasible analogues for exploring widely varying coupling regimes. For example, candidate twistronic materials can be categorized based on their intralayer and interlayer coupling strengths, then prototyped as simple phononic metamaterials by appropriately adjusting the pillar and membrane properties. This approach could inform the design of twisted vdW heterostructures with larger magic angles.

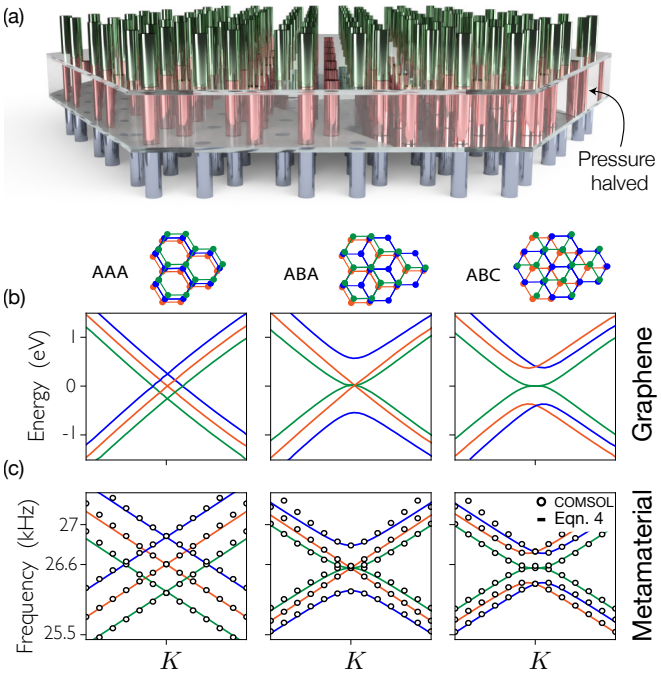

Fig. 4. **A phononic metamaterial analogue of trilayer graphene**. **(a)** In a trilayer phononic metamaterial, the boundary conditions on the inner and outer layers are distinct, creating varying acoustic intensity profiles that can be balanced by reducing the inner layer air pressure. The metamaterial comprises of three honeycomb lattices of steel pillars separated by 0.22-mm thick HDPE layers. **(b)** The Dirac cone structure of trilayer graphene displays qualitatively different behavior in each of its three different stacking configurations, adapted with permission from Ref. [30]. Copyright 2017, American Chemical Society. **(c)** Each behavior is recreated in a trilayer metamaterial by matching the stacking pattern. The calculated band structure (open circles) is well described by a simple two-parameter Hamiltonian (Eqn. 4, solid lines).

III. TOWARDS FULL VDW HETEROSTRUCTURES

9 Our interlayer coupling scheme generalizes beyond bilayer metamaterials with ease. Broadly speaking, full vdW heterostructures combine elements from a vast library of two-dimensional materials to realize emergent behavior that further diversifies as the number of layers increases. To demonstrate this trend in vdW metamaterials, we simulated an analogue of trilayer graphene by stacking three honeycomb lattices of steel pillars, separated by two HDPE membranes (Fig. 4(a)). Trilayer graphene has three possible stacking configurations, each encoded by a different interlayer hopping matrix, δ. In each case, we successfully engineered a similarly-stacked trilayer metamaterial with interlayer interactions that promote a nearly identical band structure to trilayer graphene, as shown in Fig. 4(b-c). Both the electronic and acoustic system can be described by the same tight-binding Hamiltonian, which is a simple extension of Eqn. 1,

$$\mathcal{H}(\mathbf{k}) = \begin{bmatrix} E(\mathbf{k}) & \delta_1 & 0 \\ \delta_1^T & E(\mathbf{k}) & \delta_2 \\ 0 & \delta_2^T & E(\mathbf{k}) \end{bmatrix}, \tag{4}$$

where the stacking-dependent coupling matrices are given by

$$\delta^{(1)}_{AAA} = \delta^{(2)}_{AAA} = \frac{\delta_{AA}}{\sqrt{2}} \tag{5}$$

$$\delta^{(1)}_{ABA} = \frac{\delta_{AB}}{\sqrt{2}} \qquad \delta^{(2)}_{ABA} = \frac{\delta^T_{AB}}{\sqrt{2}} \tag{6}$$

$$\delta^{(1)}_{ABC} = \delta^{(2)}_{ABC} = \frac{\delta_{AB}}{\sqrt{2}}. \tag{7}$$

As it turns out, the air pressure in the middle layer must be halved for the band structure to match that of trilayer graphene. This empirical observation presumably stems from the different boundary conditions between the inner and outer layers. Specifically, the outer boundary conditions are perfectly impedance matched, which absorbs 100% of the acoustic power, whereas the inner layer does not. Consequently, reducing the inner-layer pressure likely balances the acoustic intensity in the three layers, though it is intriguing that this behavior departs from trilayer graphene. Aside from this change, the properties of each layer, as well as the interlayer membranes, are identical to the bilayer and monolayer system (Fig. 1 and 2).

10 Unlike their quantum counterparts, vdW metamaterials can be tuned to span a continuous assortment of band structures. Although we focus primarily on analogues of graphene, parallels to other vdW materials can be easily drawn by tuning the structure and properties of the metamaterial. For example, hexagonal boron nitride is a common ingredient in many vdW heterostructures because it hosts a large insulating band gap[33]. It consists of two interpenetrating triangular lattices of boron and nitrogen, and consequently breaks the C_6 of the unit cell to open a gap at the K point. Similarly, an analogue of hexagonal boron nitride can be produced by breaking the C_6 symmetry of the graphene metamaterial in Fig. 1(c), for example, by using pillars of different radii. Unlike the electronic version, the resulting band gap in the metamaterial can be continuously tuned because the radii may be chosen arbitrarily. Metamaterials exhibiting such wide band gaps have been extensively explored in the literature[34]. Our coupling scheme allows them to be incorporated into a vdW metamaterial, affording enormous flexibility in recreating, exploring, and extending the emergent phenomena of vdW heterostructures.

IV. CONCLUSIONS

11 The essential ingredient of vdW heterostructures—their interlayer coupling—extends beyond electronic systems. By adding a flexible membrane between phononic metamaterials, we demonstrated that this coupling can be accurately recreated in acoustics. The resultant vdW metamaterials permit the rapid exploration of diverse stacking combinations and extensive coupling regimes that are challenging to reach in electronic materials. To illustrate their potential, we developed macroscopic acoustic analogues of every stacking configuration of bilayer and trilayer graphene. Our results provide a guide for designer vdW metamaterials, which may focus and inform future vdW-based electronic devices. Conversely, they permit mapping intriguing electronic phenomena to phononics, with immediate applications. Finally, our design scheme applies directly to photonic metamaterials through a simple mapping of variables.

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