Monolayer Graphene Can Emit SHG Waves

The Dynamical Centrosymmetry Breaking Mechanism

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Abstract: The usually-held notion that monolayer graphene, a centrosymmetric system, does not allow even-harmonic generation when illuminated at normal incidence is challenged by the discovery of a peculiar effect we term the dynamical centrosymmetry breaking mechanism. This effect results in a global pulse-induced oscillation of the Dirac cones which in turn produces second harmonic waves. We prove that this result can only be found by using the full Dirac equation and show that the widely used semiconductor Bloch equations fail to reproduce this and some other important physics of graphene. These results clear the way for further investigation concerning nonlinear light-matter interactions in a wide range of two-dimensional materials admitting either a gapped or ungapped Dirac-like spectrum.

Keywords: graphene, Dirac fermions, light-matter interactions, nonlinear optics, second-harmonic generation

PACS: 42.65.-k Nonlinear optics; 42.65.Ky Frequency conversion; harmonic generation, including higher-order harmonic generation; 42.65.Sf Dynamics of nonlinear optical systems; optical instabilities, optical chaos and complexity, and optical spatio-temporal dynamics; 42.65.Wi Nonlinear waveguides

1 Introduction

Graphene has been in the spotlight for more than a decade now, mostly due to its first physical realisation performed by Novoselov and Geim [1]. This two-dimensional system is truly unique – its low-energy electrons can be suitably modelled by massless Dirac fermions [2]. Their dispersion is linear in momentum, giving rise to the very famous Dirac cones. These characteristics alone allow graphene electronics to display unusual and different features from conventional semiconductor electronics [2]. Unlike most electronic properties, its optical properties are still being intensely researched and not fully understood. Similarly, novel and non-trivial features are expected [3], most notably in photonics for ultrafast photodetectors [4], optical modulation [5], molecular sensing [6] and several nonlinear applications [7, 8]. Its optical response is characterised by a highly-saturated absorption at rather modest light intensities [9], a remarkable property which has already been exploited for mode-locking in ultrafast fiber-lasers [10].

The high nonlinear response of graphene is known to lead to the efficient generation of higher harmonics [11, 12]. Theoretical approaches to model the nonlinear dynamics of graphene typically rely on the Boltzmann transport equation, accounting only for intraband electron dynamics [13] and on the popular semiconductor Bloch equations (SBEs), where both the dispersion and density of states are replaced by graphene’s.

As it will be shown in this article, the SBEs only take interband transitions into account. We show the existence of a previously unknown nonlinear optical phenomenon that occurs when a graphene monolayer is illuminated by a short and intense pulse at normal incidence.

Due to the oscillations of the Dirac cone, induced by the pulse, a breaking of the layer’s centrosymmetry takes place dynamically. This mechanism is responsible for the generation of second harmonic radiation. It will then be shown that this novel nonlinear effect can only be found by using the full Dirac equation while the SBEs completely fail to describe it. Finally, a discussion of the deep moti-
vations behind this failure in the latter equations will be included.

\[ \text{Figure 1: (colour online) Graphical depiction of the centrosymmetry breaking mechanism for normal incidence conditions. In momentum space, the field-free dispersion becomes globally shaken by the photon momentum \((e/c)A(t)\). This shift has an effect on the dynamics of a particular two-level system, here depicted by the two points in the leftmost cone set. The Dirac point - the point \(p_x = p_y = 0\) and coloured red - is dynamically shifted, leading to a breaking of the centrosymmetry \(k \leftrightarrow -k\) which in turn allows even-harmonic waves to be emitted.} \]

1.1 Quasirelativistic dynamics

The dynamics of a massless electron of charge \(-e\) and momentum \(p \equiv h\mathbf{k}\) when interacting with an electromagnetic vector potential \(A(t)\) is governed by the 2-dimensional Dirac equation \([1, 14]\):

\[ i\hbar \partial_t |\psi_k(t)\rangle = v_F \sigma \cdot \left( \mathbf{p} + \frac{e}{c} A(t) \right) |\psi_k(t)\rangle \]  

\( (1) \)

Here, \(c\) is the speed of light in vacuum, \(v_F = c/300\) is the Fermi velocity of such electrons, \(\sigma \equiv (\sigma_x, \sigma_y)\) is the 2D Pauli matrix vector and \(|\psi_k(t)\rangle\) is the time-dependent 2-spinor representing electrons in the conduction and valence bands for that specific momentum. It is worth emphasising that this 2-dimensional equation describes the dynamics of electronic states in the vicinity of the \(K\) point. Graphene admits both time-reversal and inversion symmetries and hence identical dynamics around either one of the two inequivalent Dirac points \(K\) and \(K'\), rendering the study of one such valley sufficient. More technical information can be found in \([15]\).

Normal incidence conditions are assumed; in the Coulomb gauge \(\nabla \cdot \mathbf{A} = 0\) and, without loss of generality, the electric field \(\mathbf{E} = -(1/c)\partial_t \mathbf{A}\) is assumed linearly polarised along the arbitrary \(\hat{x}\) axis, yielding \(\mathbf{A} = (A, 0, 0)\).

Due to the time dependence of the Dirac Hamiltonian, general analytical solutions of Eq. (1) are not known. However, one can seek solutions by expanding over the instantaneous band eigenfunction basis, as previously performed by Ishikawa \([14, 16]\), seek such solutions by expanding over an instantaneous band eigenfunction basis \(|\psi_{k_1}(t)\rangle \equiv (e^{-i\theta_1(t)}A, e^{i\theta_1(t)})e^{-i\theta_2(t)}/\sqrt{2}\).

These two states are labelled by the band index \(\lambda - \lambda = +1\) \((\lambda = -1)\) denotes conduction (valence) band solutions; \(\theta_1(t) \equiv \arctan(p_y/(p_x + \xi A(t)))\) is the dynamical angle and \(\Omega_k(t) \equiv (v_F/h) \int_{-\infty}^{t} (p_x + \xi A(t))^2 + p_y^2 dt^{1/2}\) is the dynamical phase. Note that both include the appropriate correction given by the photon momentum contribution. In this fashion, the field interaction is accounted for by generalising the field-free electron eigenstates to a time-dependent ansatz: \([14, 16]\)

\[ |\psi_k(t)\rangle = c_{+}(t)|\psi_{k_1}(t)\rangle + c_{-}(t)|\psi_{k_2}(t)\rangle, \]  

\( (2) \)

The dynamics of this quasirelativistic system can be made more transparent by obtaining the time derivatives of \(c_{\pm}\) and introducing new dynamical variables - the ‘population inversion’ \(w_k \equiv |c_{+}|^2 - |c_{-}|^2\) and the ‘microscopic polarisation’ \(q_k \equiv c_{\pm} e^{2i\Omega_k(t)}\), where \(\omega_0\) is the central frequency of the input pulse. Without using any approximations, the full Dirac equation (1) can consequently be recast in the following form:

\[ i\hbar \partial_t |\psi_k(t)\rangle = v_F \sigma \cdot \left( \mathbf{p} + \frac{e}{c} A(t) \right) |\psi_k(t)\rangle \]  

\( (3) \)

\[ w_k + i(2\Omega_k - \omega_0 - i\gamma_2)q_k + \frac{i}{2} w_k \partial_t k e^{i\omega_0 t} = 0, \]  

\( (3.a) \)

\[ \dot{w}_k + \gamma_1 (w_k - w_k^0) + i\partial k (q_k e^{i\omega_0 t} - q_k^* e^{i\omega_0 t}) = 0. \]  

\( (4) \)

These equations will be henceforth termed the Dirac-Bloch Equations (DBEs). In them, \(\dot{\Omega}_k \equiv (v_F/h)\int (p_x + \xi A(t))^2 + p_y^2 dt^{1/2}\) and \(\dot{\theta}_k \equiv e\omega_0 (p_x + \xi A(t))^2 + p_y^2\); \(\gamma_2\) and \(\gamma_1\) model phenomenologically the decay rates of the polarisation and population inversion, respectively. A more thorough explanation of the mechanisms underlying these decay rates is provided in subsection 1.2.

Two pivotal observations can be made when considering Eqs. (3-4). Firstly, the dipole moment that multiplies the electric field inside the function \(\dot{\theta}_k(t)\) is given by \(M_k \equiv \xi \epsilon_0 e\omega_0 (p_x + \xi A(t))^2 + p_y^2\), and it is therefore a time dependent quantity. This is very unusual in the theory of twollevel systems and we are not aware of any other realistic physical situations in which the dipole moment is temporally oscillating with the pulse.

Secondly, the frequency detuning between a specific two-level system with wavevector \(k\) and the pulse frequency \(\omega_0\) is also oscillating in time as \(2\dot{\Omega}_k - \omega_0 = \frac{2\xi \epsilon_0}{\hbar} [(p_x + \xi A(t))^2 + p_y^2] = 2\dot{\theta}_k - \omega_0\).
\[ \frac{\partial}{\partial t} A(t)^2 + \frac{p^2}{2m} \] 
In other words, the pulse itself modulates the band structure continuously, leading to global dynamical oscillations of the Dirac cone. We shall see the significance of such a modulation in the generation of new harmonics when discussing the centrosymmetry breaking effect below. These features are not present in the SBEs, since the photon momentum is neglected in that formulation.

Finally, one must acknowledge the fact that the DBEs implicitly neglect Coulomb interactions amongst the carriers. The reasons as to why such effective correlations are absent or rather small in graphene are surprising and stem primarily from QFT calculations in two dimensions [17] and renormalisation techniques of the Rabi frequency and band energies [18] (see Refs.[15, 19–21] for more information).

### 1.2 Temperature and doping considerations

The inclusion of dephasing effects are included phenomenologically through the coefficients as the inverse of the relaxation times \( \gamma_{1,2} \equiv 1/T_{1,2} \) whereas the effect of intrinsic, field-independent parameters such as temperature and chemical potential can be incorporated in a momentum-dependent equilibrium value of the populations, \( w_k^0 \). Typically, at zero temperature one has \( w_k^0 = -1 \), implying that all carriers are initially in the valence band, irrespective of their momenta. For non-vanishing temperatures and/or a non-vanishing chemical potential \( \mu \), the starting population is given by \( w_k^0 = -\sinh(x)/(\cosh(x) + \cosh(y)) \), where \( x \equiv \hbar |k| v_{\text{F}} / (k_0 T) \) and \( y \equiv \mu / (k_0 T) \). The values of such rates are dictated mostly by the substrate, impurity content and pump energy. Performing such measurements comes with its inherent difficulties. However, measurements of \( T_1 = 150 \text{ fs} \) and \( T_2 = 0.8 \text{ ps} \) for monolayer graphene have been obtained. For a more encompassing explanation of the originating mechanisms of such decays and respective measurement, we refer to [22].

It must be highlighted that in the absence of dephasing, i.e. when \( \gamma_1 = \gamma_2 = 0 \), the law of conservation of probability for each ‘two-level system’ of wavevector \( k \) (\( w_k^0 + 4/3q_k^2 = 1 \)) is also satisfied. Such an assumption is realistic for ultrashort pulses in the coherent regime, i.e. for pulse durations much shorter than the dephasing times, \( t_0 \ll T_{1,2} \), where \( t_0 \) is the input pulse duration.

### 1.3 Current analytics

The DBEs encapsulate the exact light-matter interactions predicted by the Dirac equation. Note that since the theory is two-dimensional, the current admits, in general, two components: \( J = (j_x, j_y)^T \). To extract information on the current generated by electron and hole state contributions, we proceed by determining the \( \mu \) component \( (\mu = x, y) \) of the contribution of a single momentum state in time domain – here termed a microscopic current \( j_{\mu,k} \) – by applying the current density operator \( \hat{J}_{\mu,k} = -e(\partial H_k / \partial k_\mu) = -ev_\mu \sigma_\mu \) to the ansatz \( |\psi_k\rangle \) of Eq. (2), yielding:

\[
\hat{J}_{\mu,k} = \langle \psi_k | \hat{\nabla}_{\mu,k} | \psi_k \rangle - \langle \psi_{-1,k} | \hat{\nabla}_{\mu,k} | \psi_{-1,k} \rangle,
\]

which can be computed exactly as:

\[
\begin{align*}
 j_{x,k} & = -ev_F \left( \cos \theta_k (w_k + 1) - 2 \sin \theta_k \text{Im} \left( k e^{-i\omega t} \right) \right) \\
 j_{y,k} & = -ev_F \left( \sin \theta_k (w_k + 1) + 2 \cos \theta_k \text{Im} \left( k e^{-i\omega t} \right) \right)
\end{align*}
\]

Note that the last term of Eq. (5) was added so that the current associated with occupied fermionic states of the valence band does not contribute to the total photogenerated current. This step prevents the appearance of a divergent, non-physical currents.

Finally, the observable current is obtained in the continuum limit as \( J_\mu = (g_x g_y / 4 \pi^2 d) \int \hat{J}_{\mu,k} \, dk \). Here \( g_x = g_y = 2 \) are respectively the spin (up/down) and valley (\( K / K' \)) degeneracies, \( d = 0.33 \text{ nm} \) is the monolayer’s thickness and \( dk \equiv kdkd\phi \) is the differential element in two-dimensional momentum space.

Notice that, since the electric field is linearly polarised along the arbitrary direction \( \hat{x} \), the integrated current component \( J_x \) vanishes identically.

For the \( x \) component, Eq. (6) allows the identification of intraband current (responsible of electronic transitions within the same band), proportional to \( (w_k + 1) \) and the interband current (responsible for vertical transitions between the valence and conduction bands), proportional to \(-2 \text{Im}(q_k e^{-i\omega t}) \) - hence \( J_x = J_{\text{intra}} + J_{\text{inter}} \).

For the remaining part of this article, notably subsection 2, realistic, zero-averaged localised electric and vector potential fields are assumed so not to introduce unphysical static electric fields – \( A(t) = A_0 \text{sech}(t/t_0) \sin(\omega_0 t) \) and \( E(t) = -\partial A / \partial c \).
1.4 Dynamical centrosymmetry breaking as a mechanism for second harmonic generation

The DBEs shown in Eqs. (3)-(4) must be directly compared with the well-known semiconductor Bloch equations (SBEs), widely employed in the theoretical description of the physics of gapped semiconductors [18, 23] and, most critically, put to use in simulations of light interaction with graphene [24–26].

The instantaneous energy eigenvalues derived from Eq. (1) are $\epsilon_{\lambda,\mathbf{p}}(t) = \lambda v_F \sqrt{(p_x + \xi A(t))^2 + p_y^2}$. If no external field is present, $A(t) = 0$ and the unperturbed spectrum depicted by the static Dirac cones is recovered: $\epsilon_{\lambda,\mathbf{p}} = \lambda v_F |\mathbf{p}|$. Otherwise, the whole Dirac cone oscillates around the $\mathbf{k} = 0$ point (representing the Dirac points $\mathbf{K}$ or $\mathbf{K}'$), together with the pulse along the $p_x$ direction due to the pulse polarisation along $\hat{x}$. A graphical depiction of this oscillation is shown in the sketch of Fig. 1.

The reason as to why second harmonics can be radiated from a centrosymmetric system such as graphene is related to such shift. When the Dirac cones are displaced, an inversion symmetry in momentum space, namely $\mathbf{k} \leftrightarrow -\mathbf{k}$, is temporarily broken and so is the inversion symmetry in real space $\mathbf{r} \leftrightarrow -\mathbf{r}$. These lead to a dynamical breaking of the centrosymmetry, induced by the pulse. If the pulse is intense enough, this mechanism leads to the possibility of radiating SHG radiation at normal incidence, a situation which is normally forbidden due to the supposed centrosymmetry of the graphene lattice.

Note that, since such a shift is global i.e. applied equally to all momentum states, it is far from obvious that it affects the physical current. This effect is only verified once, in principle, all momentum contributions to the current are appropriately averaged.

This phenomenon can in principle be found when analysing the effects produced on the incoming field i.e. through signatures in the reflected and transmitted fields. Rough qualitative features in time-domain may be appreciated. The transmitted field will tend to retain the form of the incident field, with a decreased amplitude. As for the reflected field, it will resemble the derivative of the incident field, with a comparatively small amplitude. The details of how both fields are affected by the optical response of graphene become very non-trivial once the incident field amplitude is high and the dynamical centrosymmetry breaking mechanism becomes non-negligible. Although this regime can in principle be calculated and implemented numerically, we have found the photo-generated electric current to be far more tractable and reliable to predict the higher-harmonic response of the material.

The dynamical centrosymmetry breaking mechanism is a previously unknown effect in graphene and is the central result of this article. It is important to distinguish conceptually this mechanism from the well-known Photon Drag Effect - a relativistic phenomenon in which the pulse is illuminated obliquely on the graphene layer – which also leads to THz and SHG effects [27, 28].

2 Simulations and implementation

The DBEs [Eqs. (3-4)] have been solved numerically with the aid of a highly accurate sixth-order Runge-Kutta algorithm. These following results were obtained with a setup wherein the graphene monolayer is pumped with a normally incident $t_0 = 10$ fsec pulse of central wavelength $\lambda_0 = 800$ nm, intensity $I = 114$ GW/cm$^2$, and at temperature $T = 0$ K and in undoped conditions. Without such dephasing effects, the conservation law alluded to in subsection 1.2 allows an adaptive method to be implemented so all numerical outputs are within a stipulated strict tolerance margin.

Figure 2(b) shows the temporal evolution of the total current $J_x(t)$. This quantity is obtained by solving the DBEs for a particular momentum state, repeating the procedure over a suitable momentum range and by finally averaging them appropriately, as explained in subsection 1.3. Note that $J_x(t) = 0$ after integration over the momenta, as it should be since the pulse is linearly polarised along the $\hat{x}$-axis. However, nothing prevents the study of arbitrary polarisation configurations of the system, yielding in general $J_y \neq 0$.

No substantial differences in time domain are found by obtaining the integrated current with either the DBEs or SBEs. However, their respective spectra reveal otherwise. In the first column of Fig. 3, [panels (a)-(b)], output spectra are shown. In Fig. 3(a), the output spectrum $S(\omega) = |\omega f(\omega)|^2$ in dB of the total current, intraband plus interband, when using the DBEs, is shown. The dynamical centrosymmetry breaking mechanism described above leads to a relatively strong SHG signal, indicated in the figure, at $\omega/\omega_0 = 2$. This SHG signal is an absolute novelty in the theory of graphene, since it was previously thought to be impossible to obtain such signal in normal incidence conditions. Figure 3(a) also shows the more conventional high-harmonic generation typical of a $\chi^{(3)}$ material, with peaks emitted at odd integer values of $\omega/\omega_0$.

Figure 3(b) shows the separated contributions of intraband (red solid line) and interband (dashed blue line) cur-
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It can be observed that, for the odd-order generation peaks, the intraband and interband currents contribute similarly to the emission as the two lines are almost superimposed. This exchangeability is not seen in even-harmonic generation – the SHG peak mainly stems from intraband current contributions.

Figures 3 [panels (c)-(d)], show the same quantities as panels (a)-(b), this time calculated by using the SBEs. We can see that no SHG is predicted by the SBEs and therefore these equations ultimately fail to show evidence of important physics contained in the Dirac equation (1). One can also notice that Fig. 3(c) and (d) are identical, since the intraband contributions are totally absent in the SBE formulation. Furthermore, the intensities of the odd-harmonic peaks in the SBEs in Fig. 2 [panels (c)-(d)] are overestimated with respect to their counterparts obtained by using the DBEs in Fig. 3 [panels (a)-(b)].

### 2.1 Role of doping and temperature on SHG

The role of a non-vanishing chemical potential $\mu$ on the output spectra in the DBEs was also investigated and depicted in Fig. 4, through the introduction of a doping parameter $z \equiv 2\mu/(\hbar\omega_0)$. When increasing $z$ from relatively small values ($z = 0.1$) to large values ($z = 1$), one can see that the SHG peak rapidly disappears. This is due to the fact that the interband transitions become progressively suppressed when increasing $z$. The influence of temperature in this model was also studied and it yielded no discernible differences when raising temperatures as high as $T = 3000 \, ^\circ\text{K}$.

### 2.2 Unsuitability of the SBEs for 2D quasirelativistic systems

Since their invention [23], the SBEs have been fantastically successful in describing the dynamics of interband transitions in semiconductors, exciton and exciton-polariton formation and spectra, and the semiconductor laser [18]. Immediately after the explosion of graphene research in recent years, the SBEs adapted to the graphene dispersion have been routinely applied in order to study the interaction between linear and nonlinear pulses with massless Dirac electrons [24]. However, it is now proved that the SBEs are often inadequate when studying gapless Dirac media like graphene and, for pulses that are short or intense enough, they will...
also fail even in the case of gapped Dirac media such as MoS$_2$, WSe$_2$ and phosphorene. In order to show precise conditions for the failure of SBEs, the model of Eqs. (3-4) is extended to a gapped layer, by inserting a mass term in Eq. (1), proportional to the energy gap $E_g$:

$$\hbar \partial_t |\psi_k(t)\rangle = \left[ \nu_F \sigma \cdot \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) + \frac{E_g}{2} \sigma_z \right] |\psi_k(t)\rangle,$$

where $\sigma_z$ is the diagonal Pauli matrix. The instantaneous energy eigenstates of Eq. (8) are $\epsilon_{\lambda, p}(t) = \lambda \nu_F \sqrt{(p_x + \frac{e}{c} A(t))^2 + p_y^2 + (E_g/2 \nu_F)^2}$. In the vicinity of the band gap centre ($p_x = p_y = 0$), the contribution of the photon momentum can be neglected only for those pulse amplitudes satisfying $|eA/c| \ll E_g/2 \nu_F$. In this case, the DBEs (3-4) are identical to the SBEs, since in this way one eliminates the time dependence of the frequency detuning and the dipole moment $M_k$.

The SBEs are therefore a valid description of light-matter interaction in gapped 2D Dirac media only when the pulse spectrum does not overlap substantially with the Dirac point or when the intensity of the pulse is not too large with respect to the gap energy. The validity of the condition above can be expressed in terms of the input pulse intensity $I \ll -I \ll I_{cr}$, where $I_{cr} \equiv \frac{1}{2} c c_0 \left( \frac{E_g \omega_0}{\nu_F} \right)^2$. If the intensity of the pulse is such that $I \geq I_{cr}$, the SBE description loses its validity. To make things worse, even for low-intensity light, short pulses satisfying the condition $\omega_0 t_0 < E_g/(\hbar \omega_0)$ will overlap too much with the Dirac point, also leading to the breaking of the validity of the SBEs. Therefore the SBEs description of gapped Dirac layers is approximately valid only if pulses are neither too short nor too intense.

The primary corollary of this condition is that for any media admitting an ungapped spectrum i.e. when $E_g = 0$, most notoriously monolayer graphene, it is in principle never possible to accurately describe light-matter interactions via the SBEs since the condition $I \ll I_{cr}$ can never be satisfied.

### 3 Discussion

#### 3.1 Experimental hints

The suppression of SHG for doped samples (as can be seen in Fig. 4) makes the use of suspended graphene adequate for probing the dynamical centrosymmetry mechanism. The experimental realisation of this effect is thus technically challenging. Recently, Kung-Hsuan Lin et al. reported the observation of SHG in exactly the same conditions presently discussed i.e. normal incidence [29]. Despite the presence of impurities in the sample, the authors ruled out this factor as the cause responsible for SHG. Instead, they attribute it to long-range curvature effects caused by transverse mechanical fluctuations of the layer which would break the centrosymmetry. However, our estimates show that such imperfections would provide exceedingly small SHG signals and be strongly temperature-dependent and mostly independent on the input pulse duration. We are convinced that our theory is the correct explanation for the phenomenon observed.

#### 3.2 Concluding remarks

In summary, a novel nonlinear optical effect is presented for monolayer graphene wherein an ultrashort and intense pulse continuously and globally shifts the Dirac cone in time domain. This shift leads to a temporary breaking of the centrosymmetry whenever the field does not vanish. This effect results in the emission of even harmonic radiation (most importantly second harmonic waves) at normal incidence to the layer. It must not, however, be mistaken for other setups wherein monolayer graphene has shown SHG emission, such as the Dynamic Photon Drag Effect. The conceptual distinction between the widely-used SBEs
and the DBEs is theoretically explained through the dynamical centrosymmetry breaking mechanism and used to demonstrate the unsuitability of the SBEs for modelling light-matter interactions in the ultrashort and intense optical regime. This effect might have already been manifested experimentally despite its inherent procedural complexities. The results and model presented provide new insights into light-matter interaction calculations for a general range of quasi-relativistic massless quantum systems in two-dimensional monolayers. Such formalism can be further generalised to study massive quasiparticles, admitting a gapped spectrum. Examples of such systems include transition metal dichalcogenides and phosphorene.

References


