Enhancing spin–orbit coupling in high-mobility graphene by introducing chiral space curvature

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Abstract

Graphene is an excellent two-dimensional materials with high-mobility and relativistic electronic linear dispersion. Its rich physical properties such as half-integer quantum Hall effect and device application potential have been continuously attracting great attention. However, light carbon atoms also imply negligible intrinsic spin–orbit coupling (SOC) strength which hinders its spintronic application. To enhance the SOC effect, we introduce a special deformation vector with chiral curvature, borrowed from the Einstein theory of general relativity, to mimic space warping and twisting. The derived Rashba type pseudospin–spin coupling locks the spin orientation of an electron with respect to its pseudospin. Combined with the original Dirac type Hamiltonian specifying the pseudospin orientation of an electron with respect to its wavevector, it lifts the spin degeneracy and paves the way for graphene-based spintronic devices. An estimate suggests that a Rashba type pseudospin–spin coupling of the order of 5 meV can be achieved in tens nanometer samples.

1. Introduction

Carbon(C) is one of the most important elements for life and referred to as the ‘king of the elements’ since it forms almost ten million compounds with other elements described to date. The carbon has an atomic configuration (1s)²(2s)²(2p)² and the four electrons in the 2nd shell are available to form covalent chemical bonds. The unique atomic configuration also makes carbon to show various allotropes. The best known allotropes are graphite, diamond, and buckminsterfullerene (C60). As an example, C60 is a zero-dimensional soccer-ball molecule composed of 60 carbon atoms in 20 hexagons and 12 pentagons [1]. The graphite can be viewed as a stack of graphene layers, single carbon sheet of honeycomb structure (hexagons), which are weakly coupled by van der Waals force [2]. Based on the graphene, various one-dimensional carbon nanotubes, resulted from rollback of graphene layer, have also been discovered [3].

Thus, understanding the physical properties of graphene is an important first step for understanding the rest of materials derived from it.

Although a number of research works have been done concerning the electronic and structural properties of graphene, no one really expects graphene to exist in a free-standing state before it was exfoliated experimentally in 2004 by Novoselov et al [4]. As early as 1946, the electronic band structure of graphene was calculated by Wallace [5]. It shows a famous relativistic linear dispersion near Fermi energy and has an unusual semimetallic behavior. As a result of electron–hole degeneracy and vanishing carrier mass near Dirac point, a distinctive half-integer quantum Hall effect (QHE) had been experimentally observed in high-quality graphene samples [6, 7]. Anomalous QHE was also proposed theoretically by Haldane in a honeycomb lattice model [8] with oppositely flux-modulated next-nearest-neighbor hopping term. Time-reversal-symmetry (TRS) breaking opens gaps on the two Dirac points and brings about a nonzero quantized Hall conductance in the absence of an external magnetic field. The impact of spin–orbit...
coupling (SOC) on the electronic linear dispersion near Dirac points was studied by Kane and Mele [9] and a novel topological insulator was predicted. Such state has a bulk insulator gap and topologically protected surface states carrying opposite-spin electrons in opposite directions. These TRS protected surface states are robust against small perturbations and exhibit a quantum spin Hall effect. Unfortunately, the SOC induced band-gap is of the order of 1.0 μeV, too small to sustain the perturbation from either thermal fluctuation or disorder effect [10, 11].

It is well known that SOC plays a vital role in various spin-related emerging quantum phenomena [12]. The correlation between electronic spin and orbital makes it possible to manipulate the electronic spin (orbital) by electric field (magnetic field). This paves the way for developing various spintronic devices such as spin current source and spin field-effect transistor related to spin Hall effect, and spin galvanic meter related to inverse spin Hall effect [13–15]. Since the SOC is so tiny in pristine graphene, one has to find a way to enhance the SOC first to make it useful in any spintronic devices. In view of its excellent carrier mobility and flexible mechanical property [4, 16], there are already enormous amount of works investigating their electronic and phononic properties under local strain deformation [17, 18]. Among others, Ando has analytically derived the SOC for mobile electrons in carbon nanotubes which is of the first order in term of atomic SOC [19]. Meyer et al showed that local deformation results in the occurrence of Landau levels induced by the pseudo-magnetic field brought about by effective gauge fields [20]. Huertas-Hernando et al have discussed the SOC for mobile electrons in curved graphene, nanotubes, and nanotubes with cap, they found that the SOC for flat graphene is of the second order while Rashba coupling and SOC for curved graphene and nanotubes are of the first order in term of atomic SOC. The largest value occurs in curved graphene and nanotubes which is of order of 0.2 K [21]. Berche et al found that Rashba SOC can be enhanced by graphene in-plane deformation [22]. Salazar et al studied chiral-induced-spin-selectivity effect in double-strand DNA model and found that the SOC for mobile electrons, which is of the first order in term of atomic SOC, can be tuned appreciably by tensile strain [23]. Varela et al [24] and Torres et al [25] have discussed the role of hydrogen bond on the Rashba SOCs in the chiral DNA and oligopeptides structures. The chiral-induced-spin-selectivity response to longitudinal deformation is consistent with the experimental observation [26]. However, all these studies concentrated mainly on the effect of strain deformation on orbital degree of freedom, where the impact on the spin degree of freedom has not been treated on equal footing. This is where the effect matters the most to spintronic application.

According to the Einstein’s theory of general relativity, the space–time around the Earth is both warped and twisted because of the huge mass and self-rotation of the Earth. As a result of geodetic effect and frame dragging effect, both the orbit-axis and spin-axis of the gyroscope circulating around the Earth are affected. Together they induce an effective SOC for macroscopic gyroscope as verified by the observations of gravity probe B [27]. This motivated us to explore the possibility to create an effective SOC of electrons by designing an artificial deformation vector in materials. In this paper, we propose a special type of deformation with chiral curvature in graphene and investigate its consequences on the electronic Hamiltonian. For the electrons near the Dirac points, the chiral curvature mimic both the space warping and twisting features. We found that such chiral curvature does result in a pseudospin–spin coupling which optimizes the orientations of electronic pseudospin and real spin with respect to wavevector and chiral curvature. In this way, spin degeneracy is lifted and effective SOC is established in addition to usual intrinsic SOC mechanism resulted from atomic SOC term. The rest of our paper is organized as follows. A special deformation vector with chiral curvature is described in section 2. The nature of the space dependent strain tensor is emphasized and an effective Hamiltonian as applied to the Dirac–Weyl equation is derived. In section 3, the effective pseudospin–spin coupling constant is estimated for a realistic graphene sample and three order of magnitude improvement over that of pristine graphene is achieved. The electronic band structures of graphene with deformation-induced pseudospin–spin coupling is presented. In addition to spin- and pseudospin-resolved band structures, the relative orientations of electronic pseudospin and spin with respect to wavevector are discussed in a great detail for each band. The information is indispensable for the spin and orbital tunings in designing spintronic and orbitronic devices. Section 4 is our conclusion.

2. Chiral curvature and effective Hamiltonian

A crystal deformation is described by a matrix transformation from a set of orthogonal unit vectors \( e_i = (x, y, z) \) to a deformed set of vectors \( e'_i = (I + u_i) e_i \) in a laboratory framework. \( I \) and \( u \) represent a unit matrix and a strain tensor, respectively. In terms of deformation vector \( u(x, y, z) \), an element of strain tensor is given by \( u_{ij} = \partial u_i / \partial j \). To distinguish the different components of a strain tensor, one usually splits the off-diagonal elements into a rhombohedral deformation part \( (r_{ij}) \) and a chiral deformation part \( (c_{ij}) \):

\[
\begin{align*}
\tau_{ij} & = (1/2)(\partial u_i / \partial j + \partial u_j / \partial i) \\
c_{ij} & = (1/2)(\partial u_i / \partial j - \partial u_j / \partial i)
\end{align*}
\]

In this paper, we concentrate on the chiral
deformation and ignore the other contributions at first. Under this approximation, the position vector \( r' \) after deformation is related to the position vector \( r \) before deformation by a simple rotation operation, 
\[ r' = r + \omega \times r, \]
with rotation angle given by \( \omega = (c_x, c_y, c_z) \). If \( \omega \) were a constant vector, a wavefunction transforms according to global homogeneous rotation of a crystal
\[ \psi(r', \sigma) = \psi(r + \omega \times r, \sigma) = e^{i \frac{\omega \cdot r}{\hbar}} \psi(r, \sigma). \]  
(1)

\( \psi(r, \sigma) \) and \( \psi(r', \sigma) \) are the spinor wavefunctions before and after a rotation operation. The total angular momentum \( J = L + S \) with \( L \) and \( S = (\hbar/2) \sigma \) denoting the orbital and spin angular momenta. Clearly, a homogeneous rotation operation does not change the physics and the electronic Hamiltonian remains the same. When it comes to an inhomogeneous rotation operation, nonlinearity brings about a space warping and twisting. For an inhomogeneous space rotation, equation (1) enables us to write the wavefunction in chirally deformed space \( r' \) in terms of the original undeformed Euclidean space \( r \). Use this relationship, one can easily derive the effective Hamiltonian in Euclidean space in terms of the Hamiltonian in chirally deformed space
\[ H_{\text{eff}}(r) = \exp \left[ -i \omega/\hbar H_0(r + \omega \times r) \exp i \omega/\hbar \right] \]
\[ \approx H_0(r) + (\omega \times r) \cdot \nabla H_0(r) + (i/\hbar) [H_0(r) \omega \cdot J - J \cdot \omega H_0(r)]. \]  
(2)

It should be emphasized that the above derivation treats the orbital and spin degrees of freedom on equal footing. The effective Hamiltonian includes corrections from both orbital and spin spaces in addition to the zeroth order Hamiltonian. The correction of strain deformation in orbital space had already been discussed extensively in several review articles [28], and the deformation-induced local on-site energy modulation and occurrence of gauge field had been considered by various authors. Thus, in this paper, we concentrate on the correction term of chiral deformation gradient on spin degree of freedom.

\[ H_{\text{eff}}(r) = H_0(r) + i \frac{\hbar}{|r|} [H_0(r) \omega \cdot S - S \cdot \omega H_0(r)]. \]  
(3)

\( H_{\text{eff}} \) recovers the original Hamiltonian when a uniform rotation operation is carried out. However, when an inhomogeneous rotation operation is imposed on a crystal, the first-order correction becomes nonzero and a chiral curvature induced pseudospin–spin coupling takes place. As an example, we choose \( \omega(r) = \frac{1}{2} \Omega \times r \) with \( \Omega \) chiral curvature. In this case, the elements of the chiral strain tensor are given by \( c_x = -\frac{1}{2}(\Omega_y z - \Omega_z y), c_y = -\frac{1}{2}(\Omega_x z - \Omega_z x), \) and \( c_y = -\frac{1}{2}(\Omega_y x - \Omega_x y) \). This space-dependent chiral strain tensor suggests that the deformed crystal is not only warped but also twisted.

In the vicinity of Dirac points near the corners of the hexagonal Brillouin zone \( (K = -K' = (4\pi/3a, 0)) \), the Hamiltonian of unstrained graphene can be written in terms of electronic wavevector \( k \) and pseudospin \( \tau \) referring to \( A(B) \) sublattices.
\[ H_0 = \hbar v_F (\mu k_x \tau_x + k_y \tau_y). \]  
(4)

\( v_F = (\sqrt{3}a/2\hbar) \) is the Fermi velocity with \( J \) the nearest neighbor hopping integral and \( a \) the lattice constant. \( \mu = \pm 1 \) refers to the two valleys near \( (K, K') \). The Hamiltonian yields a conduction-band \( (E_+(\mathbf{k}) = \hbar v_F |\mathbf{k}|) \) and valence-band \( (E_-(\mathbf{k}) = -\hbar v_F |\mathbf{k}|) \) touching exactly at the Dirac point. The chirality for the conduction-band is \( \tau \cdot k/|k| = \mu \) while that for the valence-band is \( \tau \cdot k/|k| = -\mu \). Thus, the sense of the chirality in the two valleys are interchanged. Also, the conduction- and valence-bands are two-fold spin degenerate.

When the above chiral deformation is applied to a graphene sample, the effective Hamiltonian is modified to include a correction term
\[ H_{\text{eff}} = \hbar v_F (\mu k_x \tau_x + k_y \tau_y) + \beta \left[ \mu \sigma \cdot (\bar{\Omega} \times \sigma) + \tau_y (\sigma \times \bar{\Omega}) \right]. \]  
(5)
\( \bar{\Omega} = \Omega/|\Omega| \) and \( \beta = \hbar v_F |\Omega|/4 \) is pseudospin–spin coupling constant. Equation (5) shows that zeroth order term determines the orientation of pseudospin \( \tau \) with respect to wavevector while the correction term determines the electronic spin \( \sigma \) with respect to pseudospin and chiral curvature \( \bar{\Omega} \). Together they specify the orientations of spin and pseudospin with respect to wavevector and chiral curvature for each band. In fact, a perturbation calculation involving the zeroth-order and the first-order terms yields a term quite similar to the usual Rashba SOC. As a result of pseudospin–spin coupling, the spin degeneracy for both conduction- and valence-bands are lifted.
Figure 1. The chirally deformed graphene and electronic bands around K point at $\theta = \pi/2$. (a) The deformation pattern calculated with an enlarged $\Omega = 0.2 \text{Å}^{-1}$. Arrows in the top sheet illustrate the chiral strain vector while the ones in the bottom sheet indicate the lateral deformation within graphene plane; (b) spin- and pseudospin-resolved band structures. Red arrows refer to the spin while blue arrows to pseudospin. The electronic bands are the same in the valley around $K'$ point but with pseudospin reversed.

3. Results and discussion

For the numerical calculation of electronic band structures and their wavefunctions, we take a lattice constant $a = 2.46 \text{Å}$ and the Fermi velocity $v_F = 10^6 \text{m s}^{-1}$ [2, 5]. For a moderate deformation of a crystal of the order of 4.5%, an estimated chiral curvature is $\Omega = |\Omega| \approx 3 \times 10^{-3} \text{Å}^{-1}$ for a nanometer sample of $L = 200 \text{Å}$. With these parameters one arrives at a Rashba type pseudospin–spin coupling constant $\beta_R = \hbar v_F \Omega / 4 \approx 5 \text{meV}$. This value is three orders of magnitude larger than that of pristine graphene. The values cited above will be used throughout the paper. However, the chiral deformation calculated using the above chiral curvature is too small to be visualized. We use an exaggerated value $\Omega = 0.2 \text{Å}^{-1}$ to demonstrate the visual effect of deformation.

Note that the pseudospin–spin coupling term derived above is very different from the usual Rashba SOC since it is momentum-independent. Different from the usual Rashba SOC which lifts the spin degeneracy away from the $\Gamma$-point, the constant pseudospin–spin coupling lifts the spin degeneracy right at the Dirac point. To see the correlation between the chiral deformation vector and the effect of pseudospin–spin coupling on the electronic structure of graphene, we first consider a simple case with the chiral curvature $\hat{\Omega} = \hat{e}_z$ perpendicular to graphene plane. The deformation vector is given by $u(r) = (\Omega/2)(x\hat{e}_x, y\hat{e}_y, -z^2) \ (z = 0)$. The chirally deformed landscape of graphene is shown in figure 1(a) with an enlarged $\Omega = 0.2 \text{Å}^{-1}$. The deformation vector in this case is dominated by its $z$-component. The chiral property of the deformation can be seen from $\nabla \times u$ which forms a vortex-like structure as indicated by the arrows in figure 1(a). The electronic band structures around $K$ point can be calculated analytically and four bands in the pseudospin and spin spaces are given by $E(k) = \pm \hbar v_F \sqrt{k^2 + (\Omega/4)^2 \pm \Omega/4}$. As shown in figure 1(b), the original Dirac cones with spin-degeneracy become spin-resolved under pseudospin–spin coupling. The four-fold-degeneracy states are also splitted into two singlets and one doublet in the order $-|\hbar v_F \Omega/2|, 0, 0,$ and $|\hbar v_F \Omega/2|$. Therefore, the lowest conduction-band and highest valence-band remain touched at $K$ point. The splitted conduction-bands have opposite spin orientations, so do the valence-bands. Furthermore, the original linear dispersions of graphene are changed into quadratic dispersions due to the constant pseudospin–spin coupling. The analysis on the spin and pseudospin of an eigen-wavefunction reveals rich information on their orientation with respect to the wavevector and chiral curvature. In a pristine graphene, it is well known that conduction-bands and valence-bands are classified by different pseudospin. In the valley around $K$ point, the conduction-band electrons have their pseudospins parallel to their wavevectors while valence-band electrons have their pseudospins antiparallel to their wavevectors. Once the pseudospin–spin coupling is taken into consideration, the above relationship between pseudospins and wavevectors remains unchanged. The spin-orientation is favored along $-\Omega \times \tau$ direction and disfavored along the opposite one. For this reason, the electronic spins of two splitted bands form anti-clock-wise and clock-wise vortices in $k_x-k_y$ plane, respectively. These spin patterns are closely
correlated with the chiral strain pattern illustrated in figure 1(a). Similar argument holds as well for two valence-bands. Because of the opposite pseudospin orientation in valence-bands to that of conduction-bands, the two splitted valence-bands have spin distribution patterns similar to those of conduction-bands, but with reversed order.

Above discussion applies also to the electronic properties in the valley around $K'$ point. The splitted electronic band structures are the same, but with the pseudospin orientations all reversed. It should be emphasized that although the spin degeneracies are lifted both for the conduction- and valence-bands, no band-gap is created at the Fermi energy due to the pseudospin–spin coupling. Also, the optical activities are preserved between the consecutive bands because the neighboring bands differ either in spin or in pseudospin, but absorption rate can differ dramatically because of the different spin–photon and pseudospin–photon coupling mechanisms.

A more interesting example is given when chiral curvature $\Omega$ is tilted away from the normal of graphene plane. Due to the isotropic nature of the electronic dispersion around the Dirac point, one can take $\Omega$ within $xz$-plane to facilitate discussion and without loss of generality. Using $\theta$ to denote the angle between $\Omega$ and $x$-axis, $(\Omega = (\cos \theta, 0, \sin \theta))$. The corresponding deformation vector has a form $u(x, y, z) = -(\Omega/2)(y^2 \cos \theta, -xy \cos \theta, (x^2 + y^2) \sin \theta)$. It is illustrated in figure 2(a) for $\theta = \pi/6$ with an enlarged $\Omega = 0.2 \text{Å}^{-1}$. From figure 2(a), one sees that in addition to the $z$-component of the deformation vector similar to the situation $\Omega = \hat{e}_z$ with a reduced amplitude $\Omega \sin \theta$, the deformation vector also acquires $x$-, $y$-components as indicated by the red arrows within graphene plane. As a result, the chiral strain vector $\nabla \times u$ is no longer restricted to the plane and rotational symmetry is broken both in the deformation vector as well as in the electronic bands around the Dirac point shown in figure 2(b). The four bands are analytically solved

$$E(k) = \pm \frac{\hbar v_F}{4} \sqrt{16k^2 + \Omega^2(1 + \sin^2 \theta)} \pm 2\Omega \sqrt{16(k_x^2 + k_y^2 \sin^2 \theta) + \Omega^2 \sin^2 \theta}. \quad (6)$$

Since the influence of lateral component of chiral curvature on electrons is spin-dependent, the degeneracy-point in figure 1(b) between the lower conduction-band and upper valence-band splitted into two degeneracy-points located along $k_y$-axis at $k = (0, \pm \Omega \cos \theta/4)$. The lowest energy of upper conduction-band and highest energy of lower valence-band remain at $K$ point. By expanding the electronic dispersions around their extreme energies, isotropic linear dispersion relations are recovered for the lower conduction-band and upper valence-band, but with renormalized Fermi velocity $v_F \cos \theta$. At the same time, the highest conduction-band and lowest valence-band are also affected by the pseudospin–spin coupling. Their extreme energies are shifted to $\pm (v_F \Omega/4)(1 + \sin \theta)$ and dispersion relations in the neighborhoods have anisotropic paraboloid shape. An effective mass is $m_c = \hbar \Omega/4v_F$ along $k_x$-axis and $m_p = \hbar \Omega \sin \theta/4v_F$ along $k_y$-axis.

Figure 2. The chirally deformed graphene and electronic bands around $K$ point at $\theta = \pi/6$. (a) The deformation pattern calculated with an enlarged $\Omega = 0.2 \text{Å}^{-1}$. Arrows in the top sheet illustrate the chiral strain vector while the ones in the bottom sheet indicate the lateral deformation within graphene plane; (b) spin- and pseudospin-resolved band structures. Red arrows refer to the spin and blue arrows to the pseudospin. White dashed lines represent the contours around the two minima $(0, \pm \Omega \cos \theta/4)$ along $k_y$-axis. The electronic bands are the same in the valley around $K'$ point but with pseudospin reversed.
Same as the chiral strain vector shown in figure 2(a), spin-orientation also adopts more complex structure. For the two conduction-bands, the spin in the lower one is parallel to \( \sigma = -\Omega \times k / |\Omega \times k| \) which is both perpendicular to the wavevector (pseudospin) and chiral curvature. It has the same pattern as that of chiral strain vector. While the upper one takes exactly the opposite spin configuration \( \sigma = +\Omega \times k / |\Omega \times k| \). Two valence-bands also take the same spin configurations as shown in figure 2(b), but with reversed order to that of conduction-bands. Clearly, the out-of-plane spin modulation is directly correlated with tilting the chiral curvature away from the normal direction. Our calculations showed that the electronic structures in \( K' \) valley have similar spin and pseudospin configurations but with all pseudospin reversed in figure 2(b). The complex spin configurations of the electronic structures offer an extra degree of freedom to design spin field-effect transistor.

4. Conclusion

In summary, we have proposed in this paper to use a chiral curvature in deformation vector to mimic in solids both the space warping and twisting features, borrowed from the Einstein theory of general relativity. In this way, one is able to create a geometry-induced Rashba-type pseudospin–spin coupling in materials with no atomic SOC. This can be used to enhance the SOC in materials such as high-mobility graphene samples composed of light carbon atoms. For a realistic deformation amplitude in tens nanometer samples, the pseudospin–spin coupling strength is estimated to be of the order of 5 meV, which is three to four orders of magnitude higher than that in pristine graphene. In the deformed graphenes with chiral curvature, the spin-degeneracy is lifted and original spin-degenerated Dirac cones are splitted into four separate bands with complex spin and pseudospin configurations. The spin resolved band structures paved the way for spintronic application of graphene in spin field-effect transistor and spin Hall effect related devices. The challenge for such man-made SOC lies in the implementation of deformation strain described by chiral curvature. A possible experimental setup to realize chiral space curvature is to use monolayer or thin film samples sandwiched between substrate and capping-layer with required strain configurations imposed on both sides.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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