

Pseudospin in optical and transport properties of graphene

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We show that the pseudospin being an additional degree of freedom for carriers in graphene can be efficiently controlled by means of the electron-electron interactions which, in turn, can be manipulated by changing the substrate. In particular, an out-of-plane pseudospin component can occur leading to a zero-field Hall current as well as to polarization-sensitive interband optical absorption.

Introduction. Electrons in solids demonstrate a huge variety of behavior which can be described by an appropriate effective Hamiltonian depending on the host crystal. For the charge carriers in graphene being an one-atom-thick planar sheet of carbon atoms densely packed in a honeycomb crystal lattice, the effective Hamiltonian at low energies turns out to be formally equivalent to the massless two-dimensional Dirac Hamiltonian[1, 2] $H_0 = \hbar v_0(\sigma_x k_x + \sigma_y k_y)$, where $v_0 \approx 10^6 \text{ms}^{-1}$ is the effective “speed of light”, \mathbf{k} is the two-component particle momentum, and $\sigma_{x,y}$ are Pauli matrices. In the original Dirac Hamiltonian the Pauli vector $\vec{\sigma}$ represents the spin orientation of a spin-1/2 particle which can be detected in Stern–Gerlach-like experiments. The carriers in graphene do also have additional degree of freedom known as pseudospin which is formally equivalent to the true spin of massless fermions but originates entirely from the peculiarity of the honeycomb lattice whose elementary cell has a basis of *two* atoms. As consequence the pseudospin is not linked with the internal magnetic moment of an electron and does not directly interact with the external magnetic field prohibiting Stern–Gerlach type experiments. In contrast to that, we predict in this letter situations where the pseudospin manifests itself in observable quantities and can be detected in transport as well as optical measurements on graphene Hall bars.

First of all we show that the exchange electron-electron interaction can alter the pseudospin orientation in a very broad range. In an eigenstate of H_0 the pseudospin is always parallel to the wave vector forming the well-known radial texture in the xy -plane. As we shall see shortly, the exchange interactions can turn the pseudospin texture to the out-of-plane phase with the out-of-plane angle depending on the absolute value of the particle momentum. This is due to the huge negative contribution to the Hartree–Fock ground state energy from the valence band (i. e. “antiparticle” states) which cannot be neglected in graphene because of the zero gap (i. e. zero effective mass of carriers) and large effective fine structure constant $\alpha^* = e^2/(\varepsilon \hbar v_0)$ where ε is the dielectric constant depending on the environment[3]. The exchange contribution to the ground state energy has previously been studied in both monolayer and bilayer graphene regarding properties such as the electronic compressibility[4–6] and ferromagnetism[7–9], but the importance of the in-

terplay between pseudospin and electron-electron interactions has been recognized only recently in bilayers[10].

Having established the possibility to create an out-of-plane pseudospin orientation by means of the exchange interaction, we apply the Boltzmann approach to derive the electrical conductivity tensor which turns out to have Hall components even though the external magnetic field is absent. The mechanism of this phenomenon is intimately linked to the pseudospin-momentum coupling which can be read out immediately from the Hamiltonian H_0 . Similar to the skew scattering of electrons on impurities in spin-orbit coupled systems partly responsible for the anomalous Hall effect,[11, 12] the carriers in graphene do also skew to one side of the Hall bar as long as their pseudospin has non-zero out-of-plane component. This effect has been intensively studied[13–15] assuming that the out-of-plane component occurs due to the band gap opened by spin-orbit coupling[13] which, however, seems to be weak in graphene[2]. We emphasize that neither spin-orbit coupling nor an external magnetic field is necessary to obtain a Hall current in graphene being in the pseudospin out-of-plane phase.

Experimental manifestations of the pseudospin are not limited to the electron skew scattering phenomenon but can also be seen in the interband optical absorption. Performing optical measurements on graphene[16] one can obtain direct information regarding conduction and valence band states without advanced sample processing necessary for transport investigations. Moreover, the peculiar properties discovered so far make graphene a very promising material for optoelectronic applications[17]. Optical absorption via the direct interband optical transitions in graphene has been investigated in the seminal paper[18] as well as in [19–24], but the mechanism considered there lies essentially in the two-dimensional nature and gapless electronic spectrum and does not directly involve the pseudospin orientation. Here we show that, due to the out-of-plane pseudospin orientation, the interband absorption is sensitive to the light polarization. In particular, it can be substantially reduced or enhanced as compared to its universal value $\pi e^2/\hbar c$ just by switching the helicity.

Exchange interactions. The Coulomb exchange Hamil-

tonian is given by

$$H_{\text{exch}}(\mathbf{k}) = - \sum_{\kappa'} \int \frac{d^2 k'}{4\pi^2} U_{|\mathbf{k}-\mathbf{k}'|} |\chi_{\kappa' k'}\rangle \langle \chi_{\kappa' k'}| \quad (1)$$

with $U_{|\mathbf{k}-\mathbf{k}'|} = 2\pi e^2/\varepsilon|\mathbf{k}-\mathbf{k}'|$ and $\kappa' = \pm$ being the band index. The eigenstates of $H_0 + H_{\text{exch}}$ can be formulated as $\Psi_{\mathbf{k}\pm}(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}}|\chi_{\pm k}\rangle$ with the spinors being $|\chi_{+k}\rangle = (\cos\vartheta_k, \sin\vartheta_k e^{i\varphi})^T$, $|\chi_{-k}\rangle = (\sin\vartheta_k, -\cos\vartheta_k e^{i\varphi})^T$, and $\tan\varphi = k_y/k_x$. Thus, a non-zero out-of-plane component of the pseudospin corresponds to $\vartheta_k \neq \pi/2$. To diagonalize $H_0 + H_{\text{exch}}$ the following equation for ϑ_k must be satisfied[25]

$$\begin{aligned} \hbar v_0 k \cos\vartheta_k + \sum_{\kappa'} \int \frac{d^2 k'}{8\pi^2} \kappa' U_{|\mathbf{k}-\mathbf{k}'|} [\cos\vartheta_{k'} \sin\vartheta_k - \\ - \sin\vartheta_{k'} \cos\vartheta_k \cos(\varphi' - \varphi)] = 0 \end{aligned} \quad (2)$$

where the integration goes over the occupied states. Note that the conduction and valence states are entangled, and the latter cannot be disregarded even at positive Fermi energies. Thus, in order to evaluate the integrals

$$\frac{\Delta E_{\text{tot}}}{\hbar v_0 \Lambda^3} = -\frac{2}{\pi} \int_0^1 dx' x'^2 (1 - \sin\vartheta_{k'}) - \frac{\alpha^*}{8\pi^3} \int_0^{2\pi} d\varphi \int_0^{2\pi} d\varphi' \int_0^1 dx \int_0^1 dx' x x' \frac{(1 - \sin\vartheta_{k'} \sin\vartheta_k) \cos(\varphi' - \varphi) - \cos\vartheta_{k'} \cos\vartheta_k}{\sqrt{x^2 + x'^2 - 2xx' \cos(\varphi - \varphi')}} \quad (4)$$

where we take into account spin and valley degeneracy by a factor of 4. The energy difference for $\alpha^* \sim 1$ is small because the integrand in Eq. (4) is always multiplied by x' and therefore vanishes at $x' \rightarrow 0$, but at larger x' the $\vartheta_{k'}$ gets close to $\pi/2$, and the integrand vanishes again. The Inset in Fig. 1 shows, however, that strong electron-electron interactions make the out-of-plane phase energetically preferable. The estimates of α^* for clean graphene vary from 2 (Ref. [3]) to 2.8 (Ref. [7]) and are on the borderline of the out-of-plane phase. Moreover, the presence of disorder can change this qualitative picture essentially[7].

The single-particle spectrum is given by

$$\begin{aligned} \frac{E_{\pm}(x)}{\hbar v_0 \Lambda} = \pm x \sin\vartheta_k - \frac{\alpha^*}{4\pi} \int_0^{2\pi} d\varphi' \int_0^1 dx' x' \\ \times \frac{1 \mp (\cos\vartheta_{k'} \cos\vartheta_k + \sin\vartheta_{k'} \sin\vartheta_k \cos\varphi')}{\sqrt{x^2 + x'^2 - 2xx' \cos\varphi'}}, \end{aligned} \quad (5)$$

and the group velocity can be written as $\mathbf{v}_{\pm} =$

in Eq. (2) a momentum cut-off Λ is necessary. Its value $\simeq 0.1\text{nm}^{-1}$ is usually chosen to keep the number of states in the Brillouin zone fixed[7], but our outcomes do not depend on any particular choice of Λ . Substituting $x = k/\Lambda$ we arrive at

$$\frac{4\pi x \cos\vartheta_k}{\alpha^*} = \int_0^{2\pi} d\varphi' \int_{k_F/\Lambda}^1 dx' x' \frac{\cos\vartheta_{k'} \sin\vartheta_k - \sin\vartheta_{k'} \cos\vartheta_k \cos\varphi'}{\sqrt{x^2 + x'^2 - 2xx' \cos\varphi'}}. \quad (3)$$

The momentum cut-off is obviously much larger than the Fermi momentum k_F at any reasonable electron doping, and therefore we can to set the lower integral limit to zero. There is a trivial solution with $\vartheta_k = \pi/2$ independent of k , and a non-trivial one $\vartheta(k)$ which is shown in Fig. 1 for different α^* . These two solutions represent to two phases with different total ground state energies $E_{\text{tot}}^{\text{in}}$ ($E_{\text{tot}}^{\text{out}}$) for the in-plane (out-of-plane) pseudospin phase. The difference $\Delta E_{\text{tot}} = E_{\text{tot}}^{\text{in}} - E_{\text{tot}}^{\text{out}}$ per volume reads

$(v_{\pm} \cos\varphi, v_{\pm} \sin\varphi)^T$ with v_{\pm} being

$$\begin{aligned} \frac{v_{\pm}}{v_0} = \pm \sin\vartheta_k + \frac{\alpha^*}{4\pi} \int_0^{2\pi} d\varphi' \int_0^1 dx' x' x (1 - \cos\varphi') \\ \times \frac{1 \mp (\cos\vartheta_{k'} \cos\vartheta_k + \sin\vartheta_{k'} \sin\vartheta_k \cos\varphi')}{(x^2 + x'^2 - 2xx' \cos\varphi')^{\frac{3}{2}}}. \end{aligned} \quad (6)$$

The dispersion law (5) is depicted in Fig. 2 for graphene placed on SiO_2 substrate. The exchange interactions shift the bands down to lower energies, but, most importantly, they open a gap between the valence and conduction band as soon as the system changes to the pseudospin out-of-plane one phase. The gap at $k = 0$ equals $\frac{e^2 \Lambda}{\varepsilon} \int_0^1 dx' \cos\vartheta_{k'}$. Note that the group velocity (6) vanishes at small momentum $k/\Lambda \ll 1$ as long as the system is in the out-of-plane phase corresponding to the almost flat bands close to $k = 0$ shown in the inset of Fig. 2. From now on we assume n-doping so that the Fermi energy is always higher than the bottom of the conduction band.

Zero-field Hall current. To describe the Hall conductivity due to skew scattering we utilize the semiclassical

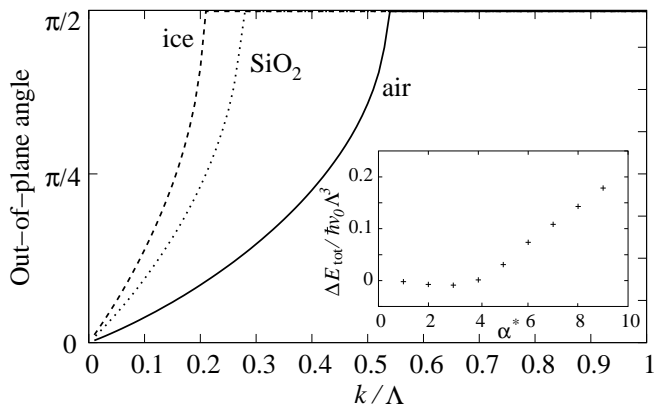


FIG. 1: The pseudospin out-of-plane angle $\vartheta(k)$ for different environments numerically calculated from Eq. (3). The corresponding values of the substrate-dependent effective fine structure constant α^* are taken from Ref. [3]. The inset shows the total ground state energy difference (4) between the in-plane and out-of-plane phases for different effective fine structure constant $\alpha^* = e^2/\varepsilon\hbar v_0$. Increasing α^* makes the out-of-plane phase more preferable.

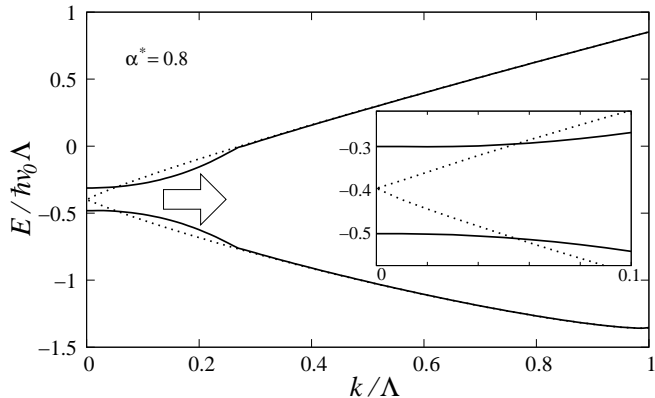


FIG. 2: The dispersion law $E_{\pm}(k)$ in the in-plane (dashed curves) and out-of-plane (solid curves) phases for $\alpha^* = 0.8$ corresponding to SiO_2 substrate[3]. The curves for both phases coincide for momenta larger than a certain critical value where $\vartheta_k = \pi/2$ becomes independent of k , see Fig. 1. The inset shows the gap region in detail.

Boltzmann approach which allows a physically transparent interpretation of this mechanism[12, 14]. In general the anomalous Hall conductivity contributions can be classified by their mechanism: (i) The intrinsic contribution is due to the anomalous velocity (being non-diagonal with respect to the band index, see Refs. [27, 28]) of carriers which is coupled to the equilibrium part of the distribution function. (ii) The side-jump contribution follows from coordinate shifts during scattering events. It occurs in the non-equilibrium part of the distribution function as well as in the anomalous velocity[12, 14]. (iii) The skew scattering contribution is independent of the coordinate shift and of the anomalous velocity. It occurs when the scattering rate is asymmetric with respect to the initial

and final states and, therefore, must be considered beyond the first Born approximation

The first two conductivity contributions do not depend on disorder but on the out-of-plane angle ϑ_k and can be adopted from [14]. The skew scattering can be described using the interband incoherent Boltzmann equation where the anomalous velocity is neglected but the scattering probability is calculated up to the third order in the short-range scattering potential which is characterized by the momentum-independent Fourier transform V . In linear order in the homogeneous electric field \mathbf{E} this equation reads $-e\mathbf{E}v_k [-\partial f^0(E_k)/\partial E_k] = I[f_{\mathbf{k}}^1]$, where $f^0(E_k)$ is the Fermi-Dirac function, $f_{\mathbf{k}}^1$ is the non-equilibrium addition, and v_k , E_k are taken from the upper cases of Eqs. (6,5). The collision integral can be written as $I[f_{\mathbf{k}}^1] = \int \frac{d^2k'}{(2\pi)^2} w_{\mathbf{k}\mathbf{k}'} (f_{\mathbf{k}'}^1 - f_{\mathbf{k}}^1)$ with $w_{\mathbf{k}\mathbf{k}'}$ being the scattering probability. We divide $w_{\mathbf{k}\mathbf{k}'}$ into two parts. The first one is proportional to the cosine of the scattering angle and calculated up to the second order in V . The second one is proportional to the sine of the scattering angle and calculated up to the third order in V . These two parts correspond to the conventional and skew scattering respectively which can be alternatively expressed in terms of the momentum relaxation times

$$\frac{1}{\tau_{\parallel}} = (1 + 3 \cos^2 \vartheta_k) \frac{n_i k V^2}{4 \hbar^2 v_k}, \quad \frac{1}{\tau_{\perp}} = \cos \vartheta_k \sin^2 \vartheta_k \frac{n_i k^2 V^3}{8 \hbar^3 v_k^2} \quad (7)$$

Here, n_i is the concentration of such scatterers. Note, that in contrast to the non-interacting case neither of τ 's diverges at $k = 0$ because of the k -dependent group velocity (6).

The solution of the Boltzmann equation can be found in the form[26–28]

$$f_{\mathbf{k}}^1 = e\mathbf{E}v_k \begin{pmatrix} a_x \cos \varphi + b_x \sin \varphi \\ a_y \cos \varphi + b_y \sin \varphi \end{pmatrix} \left[-\frac{\partial f^0(E_k)}{\partial E_k} \right] \quad (8)$$

Substituting (8) into the kinetic equation one can find easily $a_x = b_y = \tau_{\parallel}\tau_{\perp}^2/(\tau_{\parallel}^2 + \tau_{\perp}^2)$ and $b_x = -a_y = \tau_{\parallel}^2\tau_{\perp}/(\tau_{\parallel}^2 + \tau_{\perp}^2)$. Including the valley and spin degeneracy the conductivity at zero temperature reads

$$\sigma = \frac{2e^2}{h} \frac{k v_k \tau_{\parallel} \tau_{\perp}}{\tau_{\parallel}^2 + \tau_{\perp}^2} \begin{pmatrix} \tau_{\perp} & -\tau_{\parallel} \\ \tau_{\parallel} & \tau_{\perp} \end{pmatrix} \Big|_{k=k_F} \quad (9)$$

Since $\tau_{\perp} \propto 1/V^3$ whereas $\tau_{\parallel} \propto 1/V^2$ it is natural to assume $\tau_{\perp} \gg \tau_{\parallel}$, and $\sigma_{yx} \sim \sigma_{xx}\tau_{\parallel}/\tau_{\perp}$ which can vary in a quite broad range depending on the scattering parameter and Fermi momentum. In particular, at lower carrier concentrations one can approximate $\tau_{\parallel}/\tau_{\perp} \sim (k_F \vartheta_{k_F}^2 V)/(8\hbar v_{k_F})$. Note, that the Hall current changes to opposite direction if the sample becomes p-doped because the electrons and holes having antiparallel pseudospins skew to the opposite sides of the Hall bar.

Interband optical absorption. From H_0 one can deduce the following interaction Hamiltonian between the electromagnetic wave and carriers in graphene $H_{\text{int}} = \frac{ev_0}{c}(\sigma_x A_x + \sigma_y A_y)$ which couples the vector potential \mathbf{A} and pseudospin $\vec{\sigma}$. As consequence, the inter-band transition matrix elements turn out to be sensitive to the light polarization and pseudospin orientations in the initial and final states. To be specific we assume monochromatic light of frequency ω , normal incidence (i.e. zero momentum transfer from photons to electrons), and circular polarization (fulfilling $A_x = \pm iA/\sqrt{2}$, $A_y = A/\sqrt{2}$). The probability to excite an electron from the valence band to an unoccupied state in the conduction band can be calculated using the golden-rule. Finally, the absorption P can be calculated as a ratio between the total electromagnetic power W_a absorbed by graphene per unit square and the incident energy flux $W_i = \omega^2 A^2/4\pi c$. After some algebra we obtain the following expression

$$P = \frac{\pi e^2}{\hbar c} \frac{16\Lambda v_0}{\omega} \int_0^\infty dx x \left\{ \frac{\sin^4 \frac{\vartheta_k}{2}}{\cos^4 \frac{\vartheta_k}{2}} \right\} \delta \left(\frac{E_+ - E_- - \hbar\omega}{\hbar v_0 \Lambda} \right). \quad (10)$$

In the non-interacting limit the absorption equals to the universal value $\frac{\pi e^2}{\hbar c}$, as expected[18]. Note, that the integrand in Eq. (10) contains the multipliers $\sin^4(\vartheta_k/2)$ and $\cos^4(\vartheta_k/2)$ for two opposite helicities of light. At small k/Λ the absorption of the right-hand circularly polarized light is substantially reduced whereas it is facilitated in the opposite case. Moreover changing the excitation energies $\hbar\omega$ one can investigate the dependence $\theta(k)$ shown in Fig. 1. In the in-plane phase with $\vartheta = \pi/2$ the absorption does not depend on light polarization but its absolute value is renormalized due to the exchange interactions.

Conclusions. We have demonstrated that the pseudospin being until now rather uncontrollable and almost unmeasurable quantity can be “unfrozen” by the exchange electron-electron interactions (1) and play an essential role in optical and transport properties of graphene. Thus, the pseudospin can be seen as an additional degree of freedom similar to the true spin but unaffected by the magnetic field directly. We hasten to say that the Hartree-Fock approximation employed here has generically a tendency to overestimate ordering such a the pseudospin out-of-plane polarization. We believe, however, that the pseudospin eigenstates $|\chi_{\pm k}\rangle$ derived above are much more robust because their special pseudospin-momentum entangled structure stems from the free Hamiltonian H_0 , and the electron-electron interactions do only modify it. Thus, the predictions made here should be reliable at the qualitative level.

Having this similarity in mind one can think about pseudospin ferromagnetism[10], pseudospin accumulation at the sample’s edge by means of the zero-field Hall current (9), pseudospin selectivity in the optical absorp-

tion (10), and, probably, pseudospin filtering and switching. In a more distant future one can imagine some useful effects based on the pseudospin polarization like an all-electrical counterpart for GMR which is obviously very promising for application. This Letter should be seen as a first step in this direction.

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