Research Article

Gate controllable optical spin current generation in zigzag graphene nanoribbon

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Abstract

Considering the demand for long spin communication distance in spintronics, graphene presents micrometer spin relaxation length at room temperature, making it one of the most promising two-dimensional spintronic materials. However, achieving efficient spin injection (including pure spin current and spin polarized current) by reducing the spin dependent scattering between graphene and other materials like contact is still a core challenge. Here, we propose a novel approach to generate spin current in zigzag graphene nanoribbon (ZGNR) via photogalvanic (or photovoltaic) effect (PGE) from atomic first principle calculations. By designing ZGNR based device with spatial inversion symmetry, we find that the PGE induced pure spin current can be hiddenly generated without accompanying charge current. Furthermore, through applying a dual gate in the system, the generated pure spin current can be controlled when dual gate voltages have the opposite signs. Interestingly, when the signs of dual gate voltages are the same, the pure spin current can turn into the fully spin polarized current. More importantly, the generated spin current via PGE is independent of photon polarization and incident angles. Our investigations demonstrate ZGNR's great potential application in noninvasive spin injection of the graphene based spintronic device.

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1. Introduction

Nowadays, the continuous scaling down of traditional complementary metal-oxide-semiconductor (CMOS) faces a great challenge [1, 2]. On the one hand, the cost of chips is largely increased because of the required advanced facilities in the fabrication process. On the other hand, the transistor size is approaching the limit where quantum effect plays an important role. For instance, the increase of gate leakage current due to the quantum tunneling effect which in turn increases the stand by power dissipation. However, the rapid growth of the Internet of Things and big data are still in high demand for much lower power and faster speed in data process and storage. Therefore, people are resorting to other technologies beyond CMOS technology.

As one of the most promising ultra-fast and low-power consumption devices, spintronics aims to utilize the spin degree of freedom as the information carrier instead of charge itself [3]. At present, many spintronic devices with different functionalities have been developed, for instance, giant magnetoresistance, spin valve and so on [4–8]. However, it is still lacking an ideal material that can transport spin information over long distances even at room temperature. Since the discovery of graphene, two dimensional materials become an important material platform in spintronics [9–15]. As one of the most promising spintronic materials, graphene has very high mobility, negligible hyperfine interaction, and especially extremely small spin-orbital coupling strength [16–19]. It is experimentally shown that the spin relaxation length of graphene can be as long as ~30 μm at room temperature and several...
graphene devices have been demonstrated experimentally [20–23]. This unique property enables the realistic transportation and manipulation of spin information in graphene based complex device structures [22–25]. It is commonly accepted that, the generation of spin is of central importance in spintronics. At present, the spins can be injected from ferromagnetic (FM) metal into graphene. Due to the presence of an interface between FM and graphene, the spin dependent scattering may cause the low spin injection efficiency [16,26–28]. By combining h-BN and topological semimetal WTe2 and graphene, the spin injection efficiency has been enhanced [29–31]. Moreover, people are developing the nondestructive optical spin injection approaches. However, it is also a challenge due to the lack of spin dependent optical selection rules and weak optical absorption in pristine graphene [32]. More recently, it is found that the photo-excited spin polarized carrier in monolayer transition metal dichalcogenides (TMDs) can be diffused into graphene via proximity coupling effect [31–36]. It is well known, one of the central tasks in spintronics is to achieve large spin polarization and the ultimate goal is to use pure spin current without accompanying charge current [37]. Thus, following questions arise naturally: is it possible to efficiently inject the spin current (including both fully spin polarized current and pure spin current) without coupling graphene with other materials? If so, can the generated spin current be further controlled conveniently?

In this letter, we propose a novel approach to generate spin current in zigzag graphene nanoribbon (ZGNR) via photogalvanic effect (PCE) or photovoltaic effect instead of using proximity coupling effect between graphene and other materials. Based on non-equilibrium Green's function (NEGF) combined with density functional theory (DFT), we find that the pure spin current can be produced in pristine ZGNR when linearly polarized or circularly polarized light is illuminated. More importantly, the pure spin current can be obtained in a wide range of photon energy and independent of photon polarization and incident angles. Through applying the dual gate voltages in the ZGNR, the magnitude and sign of pure spin current can be both controlled. When the photon energy is fixed, the generated pure spin current can be turned on and off. More importantly, the fully spin polarized current can also be produced by changing the relative signs of the dual gate voltages.

2. Model and theoretical formalism

Firstly, the atomic structures of ZGNR with different widths are fully relaxed until the residual force on each atom is smaller than 0.001 eV/Å by using Vienna ab initio simulation package (VASP) [38,39]. In the numerical calculation, the kinetic energy cutoff is chosen as 520 eV and 10 × 1 × 1 k-point mesh is chosen for the first Brillouin-zone integration [40]. The exchange-correlation potential is taken as the generalized gradient approximation (GGA) with the Perdew-Burke-Ernzerhof (PBE) form [41]. To avoid the interaction between repeated images, the vacuum space along y and z directions are chosen to be larger than 16 Å and 15 Å, respectively. Based on the optimized atomic structures, quantum transport calculations are carried out based on non-equilibrium Green’s function (NEGF) combined with density functional theory (DFT) formalism [42–44], as implemented in the Nanodcal transport package [44]. In the self-consistent simulation, the physical quantities are expanded with the linear combination of atomic orbital (LCAO) basis at the double-ζ polarization (DZP) level; the atomic cores are described by the standard norm-conserving nonlocal pseudo-potentials [45]; the local density approximation is applied for the exchange–correlation potential. The k-point mesh is adopted as 100 × 1 × 1 for the self-consistent calculation of lead regions. The self-consistency is deemed as achieved until the monitored quantities such as every element of the Hamiltonian and density matrices differ less than 1 × 10−8 a.u. between iteration steps.

Fig. 1 shows the schematic plot of a two-probe device structure based on ZGNR. Note that dual transverse gates are applied in the system to tune the generated spin current with PGE. The linearly or circularly polarized light is shining on the central scattering region (the region of length L). In the following, we shall only consider the spin current flowing into the left lead. The spin dependent photocurrent $f_{s,t_L}^{(ph)}$ can be expressed as [46–49]:

$$f_{s,t_L}^{(ph)} = \frac{ie}{h} \left\{ \text{Tr} \left[ \Gamma_L \left[ G_{ph}^{-1} + f_L(E) \left( G_{ph}^{-1} - G_{ph}^{<} \right) \right] \right] \right\}_{s,t_L} dE,$$

where $L$ indicates the left lead and $s$ and $t$ is the spin component ($s = \uparrow, \downarrow$); $e$ and $h$ are the electron charge and the Plank’s constant; $\Gamma_L = i(\Sigma_L^r - \Sigma_L^a)$ is the linewidth function and $\Sigma_L^r = [\Sigma_L^a]^\dagger$ is the retarded self-energy due to the presence of the left lead; $G_{ph}^{-1} = G_0^{-1} - G_{ph}^{<}$ represents the lesser/greater Green’s function including electron-phonon interaction [50], where the $G_{ph}^{-1}$ is the retarded/advanced Green’s functions without photons; $f_L(E)$ is the Fermi-Dirac distribution function of the left lead. The polarization of the light can be defined by a complex vector $e$. The corresponding unit vectors $e_{\uparrow}$ and $e_{\downarrow}$ are in the x and y directions respectively [as shown in Fig. 1(a)]. For a linearly polarized light, $e = \cos \theta e_{\uparrow} + \sin \theta e_{\downarrow}$. For an elliptically polarized light, $e = \cos \phi e_{\uparrow} \pm \sin \phi e_{\downarrow}$ [48]. For simplicity, we introduce a normalized photocurrent [47,50], i.e. spin related photoresponse $R_s$,

$$R_s = \frac{f_{s,t_L}^{(ph)}}{e a_0},$$

where the unit of $R_s$ is $a_0^2$/photon and $a_0$ represents the Bohr radius; $f_{s,t_L}^{(ph)}$ is the spin related photocurrent defined in Eq. (1); $e$ is the electron charge; $I_L$ is the photon flux defined as the number of photons per unit time per unit area. Thus, the charge current and spin current can be defined as

$$I_c = I_{\uparrow} + I_{\downarrow},$$
$$I_s = I_{\uparrow} - I_{\downarrow}.$$

Here, the $R_{s,1}$ represents the photoresponse with spin up or spin down component. Furthermore, the spin polarization (SP) is introduced

$$SP(\%) = \frac{||R_{\uparrow}|| - ||R_{\downarrow}||}{||R_{\uparrow}|| + ||R_{\downarrow}||} \times 100.$$

3. Results and discussion

In the beginning, we investigate the bulk electronic properties of ZGNR. Since the width of the ZGNR can be classified by the number of zigzag chains along the y direction, we refer to the ZGNR as n-ZGNR in the following (here, we mainly take 5-ZGNR as an example to analysis (if not specified)). The band structure of 5-ZGNR is shown in the left column of Fig. 2(a). It is clear that the spin up and spin down bands are degenerate. By calculating the spin density $\rho_s = \rho_{\uparrow} - \rho_{\downarrow}$ [right column in Fig. 2(a)], we can know that the charge density of spin up and down components mainly distribute at the upper and lower edges, which indicates that ZGNR is in the antiferromagnetic insulating ground state. When the external transverse gate voltage is applied [shown in Fig. 2(d)], the spin up and down bands split as

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shown in Fig. 2(b and c). Furthermore, the spin up band gap increases/decreases while the spin down band gap decreases/increases when the gate voltage is positive/negative. As the gate voltage is further increased, the band gap closes and ZGNR becomes halfmetal in Fig. 2(c). In Fig. 2(d), the changes of spin density induced by gate voltages $\Delta \rho_s = \rho_s(V_g = \pm 8 V) - \rho_s(V_g = 0 V)$ are presented. Physically, it is known that both the spin up and spin down charge densities move upward/downward under a positive/negative gate voltage. As shown in Fig. 2(d), we can know that the spin density difference is positive/negative in the lower/upper edge regardless of the gate voltages sign. However, for the positive case (left column in Fig. 2(d)), the magnitude of the negative part in the upper edge is larger than that of the negative part in the lower edge, which means the increase of spin density in the upper edge is larger than the decrease of spin density in the lower edge. This is different for the negative case. From the right column in Fig. 2(d), it is found that the increase of spin density in the upper edge is smaller than the decrease of spin density in the lower edge. This means that the transverse gate can be used as an effective tool to tune the spin-dependent electronic property of ZGNR \cite{51,52}. The spin-dependent bandgap versus the gate voltage for $n$-ZGNR ($n = 3, 5, 7, 9$) are presented in Supplement Information (Fig. S1). Based on these unique properties of the ZGNR, we construct a ZGNR based two-probe devices with dual gates as shown in Fig. 1(a). Note that the device structure holds inversion symmetry with a inversion center shown in Fig. 1(b). During the numerical simulation, the device can be divided into three parts as shown in Fig. 1(b): the central scattering region and left/right lead which extends to infinity. Here, the light (linearly polarized or circularly polarized) is vertically illuminated in the central scattering region. In the following, the left gate voltage is fixed as positive, while that exerted on the right lead can be switched. For convenience, the positive-positive and positive-negative gate voltages in the dual gate are denoted by PP and PN in the following, respectively.

We now analyze the spin current generated by linearly polarized photogalvanic effect (LPGE) and circularly polarized photogalvanic effect (CPGE). As shown in Fig. 3(a), the pure spin current is...
produced for both LPGE and CPGE in ZGNR without applying the gate voltage \( (V_g = 0 \text{ V}) \). Since the device structure maintains spatial inversion symmetry, the charge current cannot be generated by PGE. This is consistent with phenomenological theory [53]. However, the pure spin current can still be obtained if the spin density has no spatial inversion symmetry regardless of light’s polarization and its incident angle [54,55]. As shown in Fig. 2(a), the charge density with spin up or down component are distributed in upper or lower edge, respectively. Thus, the spin density is anti-symmetrically distributed \([\rho_1(\mathbf{r}) = \rho_1(-\mathbf{r})]\) and thus it is an anti-ferromagnetic coupling between any two atoms with respect to the inversion center. Compared with LPGE, the pure spin current is also obtained via CPGE while their magnitudes are much larger.

By analyzing the effective transmission (shown in Fig. S2 in SI), we can find that the transport probabilities of outgoing electrons in CPGE are much larger than that of incoming electrons in CPGE resulting in a relatively large pure spin current when \( V_g = 0 \text{ V} \). In order to know the gate voltage effect, the numerical results of spin current generated in PN and PP configurations when \( V_g = 8 \text{ V} \) are presented in Fig. 3(b and c). In PN configuration, the pure spin current can be induced when the photon energy is larger than 0.2 eV. This indicates that the pure spin current can be tuned by adjusting the gate voltage. Interestingly, the magnitude of pure spin current with LPGE is largely increased compared with zero gate voltage case (the maximum magnification factor can be as large as \( \sim 10^3 \)). This can be understood by analyzing the effective transmission shown in the first section of the Supplementary Information. While in CPGE, the magnitude of pure spin current does not change too much when the gate voltage is applied. Accordingly, the spin polarization of the PN configuration is always equal to zero in the given photon energy range shown in Fig. 3(d). In PP configuration, the spin density is no longer anti-symmetrically distributed \([\rho_1(\mathbf{r}) = \rho_1(-\mathbf{r})]\), which can be estimated from the spin density difference shown in left column of Fig. 2(d). From Fig. 3(c), we can easily find that the spin polarized current is generated with LPGE and CPGE, since the accompanying charge current is generated. Interestingly, the charge current and spin current have equal magnitude and opposite signs in a wide range of photon energies (the photon energy is less than 0.45 eV), which means \( I_c + I_s = 0 \) and the fully spin polarized current with spin down component is produced. Correspondingly, the spin polarization of PP configuration is 100% when the photon energy is less than 0.45 eV as shown in Fig. 3(d). Therefore, by tuning the dual gate voltages, the pure spin current and fully spin polarized current can both be generated in our proposed device. Moreover, the sign of pure spin current can be changed and fully spin polarized current with different spin component can be obtained by tuning the sign of dual gate voltages (See more numerical results in section II of the Supplementary Information).

To further understand the physics behind the generation of spin current, we take LPGE in PN configuration as an example to give a physical picture. In the first place, the local density of states (LDOS) of the central scattering region is presented in Fig. 4(a). Due to the presence of positive and negative gate voltages exerted on the left and right sides of the central region, the spin down and up band gap close correspondingly in these two regions and they still keep open in the other regions. In the light illumination region [between the two pink vertical lines shown in Fig. 4(a)], the valence band of spin up and down components gradually bend upward and downward, respectively, while the situation is reversed for the conduction band. Fig. 4(b) presents a qualitative physical picture of the generation of pure spin current, which can be divided into three processes. Firstly, the electron below the Fermi level with energy \( E_{\text{in}} \) can be excited into states with energy \( E_{\text{out}} = E_{\text{in}} + E_{\text{ph}} \) by absorbing incident photons \((E_{\text{ph}} = h\nu)\) in the central scattering region. From LDOS shown in Fig. 4(a), we can know that both spin up and down states with energies \(-0.4eV < E_{\text{in}} < 0\) are available to be excited. Secondly, the excited electrons will flow out of the central scattering region into the leads. It is worth mentioning that the excited electrons with spin up component can only flow into the right lead when the photon energy is not too large, since there is no available spin up states (with finite band gap) in the left lead [see Fig. 4(a)]. At the same time, the excited electrons with spin down component can only flow into left lead, since there is no available spin down states (with finite band gap) in the right lead. Thus, the minimum required photon energy to generate spin current is determined by the energy gap \( E_{\text{g1}} \) shown in Fig. 4(a and b). Thirdly, the electrons with energy \( E_{\text{in}} \) in the leads flow into the central scattering region to fill the holes left during the excitation process. Due to the presence of the spatial inversion symmetry, which means motion of incoming and outgoing electrons will cancel each other resulting in zero charge current. However, the hidden net spin flow can still be produced during this process. By rewriting Eq. (1) (where we have assumed that the temperature is low enough and the Fermi
the system is irradiated by a linearly polarized light with 
mission $T$ and the total effective charge transmission $T_{\text{eff}}$, we
found that the spin channel $T_{\text{eff}}$ can be obtained, which indicates that the pure spin current can be obtained. The similar analysis on fully spin polarized current generated in PP configuration is presented in section III of the Supplementary Information.

Having understood the physical picture of pure spin current generation, we now calculate the spin current versus the photon polarization angles by fixing the photon energy $E_{\text{ph}} = 0.4$ eV. We consider PN and PP configurations with LPGE by varying $\theta$ and CPGE by varying $\phi$, respectively. Fig. 5(a and b) plot the calculated spin current versus $\theta$ and $\phi$. In PN configuration, the pure spin current without accompanying charge current is generated and independent of polarization angles both in LPGE and CPGE cases. In PP configuration, the magnitudes of charge current and spin current are equal to each other but with opposite signs, i.e., $I_c + I_s = 2R_T$. Thus, only the current with the spin down component is obtained, which is also independent of photon polarization angles. More importantly, the magnitudes of pure spin current in PN and fully spin polarized current in PP can be largely tuned via photon polarization angles. Actually, the pure spin current and fully spin polarized current are also independent of photon incident angles (see more numerical results in the fourth section of the Supplementary Information).

Last but not the least, we study the effect of gate voltage and ZGNR's width in producing the pure spin current in PN configuration. To illustrate it, the phase diagram of pure spin current versus photon energy $E_{\text{ph}}$ and gate voltage $V_g$ is shown in Fig. 6(a). It is clear that the pure spin current can be generated in a wide range of parameter space ($E_{\text{ph}}, V_g$). Moreover, the required minimum photon energies $E_{\text{ph}}$ to generate pure spin current at different gate voltages are also marked using a black dotted line in Fig. 6(a). This indicates

![Fig. 4](image)

**Fig. 4.** (a) The local density of states (LDOS) of spin up (left column) and spin down (right column) components versus electron energy and transport direction (x). The light illumination region is between the two pink vertical lines. Physical process of the generation of the pure spin current in PN configuration is schematically plotted in (b). Here blue/red lines denote motion of spin up/down electrons, respectively. Wiggled lines denote motion of incoming and outgoing electrons. The Fermi level $E_f = 0$. (c) The effective transmission $T_{\text{eff}}$ versus electron energy $\epsilon$, 4. (d) The net effective transmission $T_{\text{eff}}$ versus electron energy $\epsilon$. Here, the gate voltage $V_g = 8$ V and the system is irradiated by a linearly polarized light with $E_{\text{ph}} = 0.4$ eV. (A colour version of this figure can be viewed online.)

![Fig. 5](image)

**Fig. 5.** Charge current ($I_c$) and spin current ($I_s$) versus polarization angle $\theta$ with LPGE (left column) and CPGE (right column), respectively. (a) PN configuration. (b) PP configuration. The blue hollow circle line and the red hollow square line represent charge current and spin current, respectively. (c) Spin polarization ($SP(\%)$) versus $\theta$ and $\phi$ for PN and PP configurations. Here the photon energy and gate voltage is fixed as $E_{\text{ph}} = 0.4$ eV; $V_g = 8$ V. (A colour version of this figure can be viewed online.)
that the pure spin current can be turned off by either tuning the photon energy or gate voltage in the system. As shown in Fig. 6(b), we find that the photon energy range of generating nonzero pure spin current gradually increases with the increasing gate voltage. In addition, the pure spin current can also be obtained in other ZGNR systems [see Fig. 6(c)], which presents a similar trend. Note that the minimum photon energy of generating pure spin current also decreases with the increase of the ribbon width due to the decreasing band gap of the corresponding system.

**4. Conclusion**

In summary, we theoretically propose a novel spin current injection approach into graphene. Here, the pure spin current and fully spin polarized current can be obtained and controlled via PGE by applying a dual gate in ZGNR based device. When the system is in PN or PP configuration, the pure spin current or fully spin polarized current can be obtained in a wide range of parameter space $(E_{ph}, V_g)$. More importantly, the generated spin current can be largely tuned by the photon energy, polarization angles and gate voltages, which means that it can be efficiently controlled. In addition, the photon energy range to produce spin current can be modulated by changing the gate voltage and nanoribbon’s width. Since the proposed device can be easily contacted with different graphene based spintronic devices, our work provides a novel noninvasive spin current injection mechanism.

**CRediT authorship contribution statement**

Liwen Zhang: Numerical Simulation, Visualization, Design the project, Supervision, Writing – original draft. Jun Chen: Design the project, Supervision, Writing – review & editing. Fuming Xu: Visualization, Writing – review & editing.

**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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**Appendix A. Supplementary data**

Supplementary data to this article can be found online at https://doi.org/10.1016/j.carbon.2020.11.033.

**References**

[1] International Roadmap for Devices and Systems: beyond Cmos, IEEE.