Abstract. To understand the main spin relaxation mechanism in graphene, we investigate the spin relaxation with random Rashba field (RRF) induced by both adatoms and a substrate using the kinetic spin Bloch equation approach. The charged adatoms, on the one hand, enhance the Rashba spin–orbit coupling locally and, on the other hand, serve as Coulomb potential scatterers. Both effects contribute to spin relaxation limited by the D’yakonov–Perel’ (DP) mechanism. In addition, the RRF also causes spin relaxation by spin-flip scattering, manifesting itself as an Elliott–Yafet (EY)-like mechanism. Both mechanisms are sensitive to the correlation length of the RRF, which may be affected by environmental parameters such as electron density and temperature. Fitting and comparing the experiments of the Groningen group (Józsa et al 2009 Phys. Rev. B 80 241403(R)) and the Riverside group (Pi et al 2010 Phys. Rev. Lett. 104 187201; Han and Kawakami 2011 Phys. Rev. Lett. 107 047207), which show either DP (with the spin relaxation rate being inversely proportional to the momentum scattering rate) or EY-like (with the spin relaxation rate being proportional to the momentum scattering rate) properties, we suggest that the DP mechanism dominates the spin relaxation in graphene. The latest experimental finding of a nonmonotonic dependence of spin relaxation time on diffusion coefficient by Jo et al 2011 (Phys. Rev. B 84 075453) is also well reproduced by our model.

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## 1. Introduction

Graphene, a two-dimensional allotrope of carbon with a honeycomb lattice, has attracted much attention in recent years owing to its two dimensionality, Dirac-like energy spectrum and potential for all-carbon-based electronics and spintronics [1–8]. With breaking of inversion symmetry, possibly caused by ripples [9], perpendicular electric fields [9–13], adsorbed adatoms [13–15], the substrate [16–18], etc, the Rashba spin–orbit coupling [10, 11, 19] arises, which results in spin relaxation in the presence of scattering in graphene. A number of experiments on spin relaxation in graphene on SiO$_2$ substrate are available, revealing a spin relaxation time of the order of 10–100 ps [3, 20–27]. However, somewhat contradictory results are obtained from in these experiments. While a decrease of the spin relaxation rate with increasing momentum scattering rate has been observed by the Riverside group by surface chemical doping at 18 K [24], a linear scaling between the momentum and spin scattering has been observed by both the Groningen group [20, 22, 23] at room temperature and very recently the Riverside group [26] at low temperature ($\lesssim$100 K) via varying the electron density in graphene. The D’yakonov–Perel’ (DP) [28] mechanism was shown to be important by the former phenomenon; however, the Elliott–Yafet (EY) [29] mechanism was suggested to account for the latter. In addition, at a temperature as low as 4.2 K, a nonmonotonic dependence of spin relaxation time on diffusion coefficient with an increase of electron density was reported very

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recently by Jo et al [27]. They claimed that the spin relaxation is due to the EY mechanism. To understand the spin relaxation in graphene fully, theoretical studies are in progress [15, 18, 30–34].

Theoretically, the EY mechanism has been revealed to be invalid to account for the linear scaling between the momentum and spin scattering with an increase of electron density [33] and also less important than the DP mechanism [30]. According to [30] and [33], the spin relaxation times caused by the EY and DP mechanisms are \( \tau_s^{EY} \approx (h v_f k_f / \lambda)^2 \tau_p \) and \( \tau_s^{DP} \approx h^2 / (\lambda^2 \tau_p) \), respectively. Here \( v_f \) and \( k_f \) are the Fermi velocity and momentum, \( \lambda \) is the Rashba spin–orbit coupling strength and \( \tau_p \) is the momentum relaxation time. Consequently, \( \tau_s^{EY} / \tau_p \sim k_f^2 \), disagreeing with the electron density-independent linear scaling between \( \tau_s \) and \( \tau_p \) in the experiments [20, 22, 23, 26]. The latest experiment by Jo et al reveals the relation \( \tau_s^{EY} / \tau_p \sim k_f^2 \) with an increase of carrier density and hence suggests that the EY mechanism dominates spin relaxation in graphene [27]. Nonetheless, from the above theoretical results it is seen that \( \tau_s^{EY} / \tau_s^{DP} \approx (k_f l)^2 \) with \( l \) being the mean free path. Typically \( (k_f l)^2 \gg 1 \) in graphene, meaning that the DP mechanism dominates over the EY one [30]. For the above reasons, the EY mechanism cannot dominate spin relaxation in graphene.

The DP mechanism thus becomes a reasonable candidate for the dominant spin relaxation mechanism. Initially, the spin relaxation time limited by the DP mechanism was calculated to be much longer than the experimental data \( \sim 10–100 \) ps due to the weak Rashba field. For example, ripples with curvature radii \( \sim 50 \) nm induce the local Rashba spin–orbit coupling with \( \lambda \sim 8.5 \) \( \mu \)eV [9], and a perpendicular electric field of magnitude \( E_z \) \( (E_z \sim 0.1 \) V nm\(^{-1}\)\) contributes a Rashba spin–orbit coupling with \( \lambda \sim \zeta E_z \), where \( \zeta \approx 0.258 \) \( \mu \)eV (V nm\(^{-1}\)) \(^{-1}\) from rough estimation [10]. 17.9 or 66.6 \( \mu \)eV (V nm\(^{-1}\)) \(^{-1}\) from the tight-binding model [9, 11] and 5 \( \mu \)eV (V nm\(^{-1}\)) \(^{-1}\) from the first-principles calculation [12, 13]. Based on this weak Rashba spin–orbit coupling, Zhou and Wu calculated the spin relaxation in graphene on SiO\(_2\) substrate with mobility \( \sim 10^4 \) cm\(^2\) (V s\(^{-1}\)) \(^{-1}\) taken from the charge transport measurement [1] and obtained quite a long spin relaxation time of the order of \( \mu s \) [31]. However, for the nonlocal measurements of the spin relaxation, the mobility is at least one order of magnitude smaller [20–23, 26, 27], most likely caused by the extrinsic factors induced by the ferromagnetic electrodes [35], e.g. the adatoms. The adatoms, as well as the influence of the substrate, may substantially enhance the Rashba spin–orbit coupling by distorting the graphene lattice and inducing \( sp^3 \) hybridization, leading \( \lambda \) to be of the order of meV [13–15, 17]. With this enhanced Rashba spin–orbit coupling, the spin relaxation time in graphene is estimated [18] and calculated [34] to be comparable to the experimental data. Nevertheless, whether and how the DP mechanism accounts for the experimentally observed linear scaling between the momentum and spin scattering is questionable.

Apart from the above two mechanisms, another EY-like mechanism may contribute to the spin relaxation in graphene when the Rashba field is random in the real space. As proposed by Sherman for semiconductors ([36, 37]); for a review, see [38]), the randomness of spin–orbit coupling contributes to or even dominates spin relaxation by spin-flip scattering under certain conditions [32, 36–39]. For graphene, the Rashba field induced by a fluctuating electric field from ionized impurities in the substrate or ripples is indeed random in the real space. The former case, with the average Rashba field being nonzero, has been investigated by Ertler et al [18] via Monte Carlo simulation, while the latter case, with the average Rashba field being
zero, has been studied by Dugaev et al [32] via the kinetic equations. However, for both cases the calculated spin relaxation time is much longer than the experimental data.

In this work, we investigate spin relaxation in graphene with random Rashba field (RRF) induced by adatoms and a substrate by means of the kinetic spin Bloch equation (KSBE) approach [40]. A random Rashba model is set up, where the charged adatoms, on the one hand, enhance the Rashba spin–orbit coupling locally and, on the other hand, serve as Coulomb potential scatterers. Based on this model, an analytical study on spin relaxation with RRF is performed. It is found that while the average Rashba field leads to spin relaxation limited by the DP mechanism, which is absent in the work of Dugaev et al [32], the randomness causes spin relaxation via spin-flip scattering. With an increase of adatom density, the spin relaxation caused by the spin-flip scattering due to the RRF always shows EY-like behaviour (the spin relaxation rate is proportional to the momentum scattering rate), whereas the DP mechanism can exhibit either the EY- or DP-like (the spin relaxation rate is inversely proportional to the momentum scattering rate) one. When all other parameters are fixed, with an increase of electron density the spin relaxation rates due to both mechanisms increase; nevertheless, the spin relaxation rate determined by the spin-flip scattering due to the RRF is insensitive to temperature, whereas that determined by the DP mechanism becomes insensitive to temperature when the electron-impurity scattering is dominant. However, the correlation length of the RRF may vary with the electron density as well as temperature and both mechanisms are sensitive to the correlation length.

We carry out numerical calculations and fit the experiments of the Riverside [24, 26] and Groningen [23] groups. Fitting the DP-like behaviour with an increase of adatom density observed by the Riverside group [24], we find that only when the DP mechanism is dominant can the experimental data be understood and that the effect of the spin-flip scattering due to the RRF is negligible. However, the experimental EY-like behaviour with an increase of electron density first observed by the Groningen group [23] can be fitted from our model with either the DP mechanism or the spin-flip scattering due to the RRF being dominant by taking into account the decrease of the correlation length of the RRF with increasing electron density. Nevertheless, the fact that the Riverside group has also observed similar EY-like behaviour in their samples very recently [26] suggests that the DP mechanism is dominant but exhibits EY-like properties with increasing electron density. The temperature dependence of the spin relaxation from the Riverside group [26], suggested to be evidence for the EY mechanism, is also fitted by our random Rashba model with the DP mechanism being dominant. The corresponding temperature dependence of the spin-flip scattering due to the RRF is demonstrated to be, in fact, temperature insensitive. The similar experimental phenomena observed with the variation of electron density by the two groups further suggest that the DP mechanism also dominates the spin relaxation in the experiment of the Groningen group [23]. Moreover, the nonmonotonic dependence of spin relaxation time on diffusion coefficient very recently reported in [27] is also fitted.

This paper is organized as follows. In section 2, we present the model and introduce the KSBEs. In section 3, we investigate the spin relaxation analytically and discuss the relative importance of the DP mechanism and the mechanism of spin-flip scattering due to the RRF. In section 4, we perform numerical calculations and fit the experimental data. We discuss the results and conclude in section 5.
2. The model and the kinetic spin Bloch equations

The $n$-doped graphene monolayer under investigation lies on the SiO$_2$ substrate perpendicular to the $z$-axis. The random Rashba spin–orbit coupling reads [10, 11]

$$H_\text{R} = \lambda(r) (\mu \tau_x \sigma_y - \tau_y \sigma_x).$$  

(1)

Here $\mu = \pm 1$ labels the valley located at $K$ or $K'$. $\tau$ and $\sigma$ are the Pauli matrices in the sublattice and spin spaces, respectively. The position-dependent coupling strength $\lambda(r)$, mainly contributed by the randomly distributed adatoms and also possibly by the substrate, can be modelled as

$$\lambda(r) = \lambda_0^1 + \sum_{n=1}^{N_4} \delta_n e^{-|r-R_n|^2/2\xi^2}.$$  

(2)

Here, the second term is contributed by the adatoms with a total number $N_4$. In this model it is assumed that an adatom located at $R_n$ induces a local Rashba field peaking at $R_n$ with a magnitude of $\delta_n$ and decaying within a length scale $\sim \xi$ following the Gaussian form. $\delta_n$ is of the order of meV ([13–15]) and $\xi$ is larger than the graphene lattice constant of 0.25 nm [15]. The first term $\lambda_0^1$ comes from the average contribution from the substrate, whose fluctuation is phenomenally incorporated by affecting $\xi$. The mean value of $\lambda(r)$ reads $\lambda_0 \equiv \langle \lambda(r) \rangle = \lambda_0^1 + \lambda_0^2$ with $\lambda_0^2 = 2\pi n_i^2 \delta^2 \xi^2$, where $n_i$ is the areal density of adatoms. The correlation function $C(r) \equiv \langle |\lambda(r) - \lambda_0| |\lambda(0) - \lambda_0| \rangle = \pi n_i^2 \delta^2 \xi^2 e^{-r^2/4\xi^2}$, with the corresponding Fourier transformation

$$C_q = 4\pi^2 n_i^2 \delta^2 \xi^2 e^{-q^2/4\xi^2}.$$  

(3)

This expression is similar to that given in [32] except that it depends on $\xi$ at a higher order here. For single-sided adatoms (the adatoms are distributed on the graphene surface), we choose $\delta_n = \delta$, from which $\bar{\delta} = \delta$ and $\overline{\delta^2} = \delta^2$. For double-sided adatoms (the adatoms are distributed both on the graphene surface and at the graphene/substrate interface), we set $\delta_n = \delta_1$ or $\delta_2$ ($\delta_1 \delta_2 < 0$) with equal possibilities, from which $\bar{\delta} = 0.5(\delta_1 + \delta_2)$ and $\overline{\delta^2} = 0.5(\delta_1^2 + \delta_2^2)$. It is noted that the random Rashba model proposed here is modified from the short-range random potential model depicting the electron–hole puddles in graphene [41].

Under the basis laid out in [18] and [31], the electron Hamiltonian can be written as [31]

$$H = \sum_{\mu, k, s} \left[ \varepsilon_k \delta_{ss'} + \Omega_k \cdot \sigma_{ss'} \right] c_{\mu k s}^\dagger c_{\mu k s'} + H_{\text{int}}$$  

(4)

in the momentum space. Here $c_{\mu k s}$ ($c_{\mu k s}^\dagger$) is the annihilation (creation) operator of electrons in the $\mu$ valley with momentum $k$ (relative to the valley centre) and spin $s$ ($s = \pm \frac{1}{2}$). $\varepsilon_k = \hbar v_f k$ with $v_f = 10^6$ m s$^{-1}$. The effective magnetic field $\Omega_k$ from the average Rashba field is

$$\Omega_k = \lambda_0 (-\sin \theta_k, \cos \theta_k, 0),$$  

(5)

where $\theta_k$ is the polar angle of the momentum $k$. The Hamiltonian $H_{\text{int}}$ consists of spin-conserving scattering (electron-impurity [42] (here the impurities include both the ones existing in the substrate and the charged adatoms [24, 43]), taken into account by the minimal model proposed by Adam and Das Sarma [42]), electron–phonon [44–46] and electron–electron [31] scattering) as well as spin–flip scattering due to the RRF [32, 38],

$$H_{\text{flip}} = \sum_{\mu, k \neq k, s s'} \lambda_{k-k'} V_{kk's'} c_{\mu k s}^\dagger c_{\mu k s'},$$  

(6)

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where
\[ \lambda_q = \int \lambda(r) e^{-iq \cdot r} dr \] (7)
and
\[ V_{kk'} = \begin{pmatrix} 0 & -ie^{-i\theta_k} \\ ie^{i\theta_k} & 0 \end{pmatrix}. \] (8)
The KSBEs are [40]
\[ \partial_t \rho_{\mu k}(t) = \partial_t \rho_{\mu k}(t) |_{coh} + \partial_t \rho_{\mu k}(t) |_{scat} + \partial_t \rho_{\mu k}(t) |_{coh}^{\prime}. \] (9)
Here \( \rho_{\mu k}(t) \) represent the density matrices of electrons with relative momentum \( k \) in valley \( \mu \) at time \( t \). The coherent terms \( \partial_t \rho_{\mu k}(t) |_{coh} = -\frac{i}{\hbar} [\mathbf{\Omega}_{k} \cdot \mathbf{\sigma}, \rho_{\mu k}(t)] \) with the Hartree–Fock term from the Coulomb interaction being neglected due to the small spin polarization [31, 40]. The concrete expressions for the spin-conserving scattering terms \( \partial_t \rho_{\mu k}(t) |_{scat} \) can be found in [31].

When the electron mean free path \( l \) is much longer than the correlation length of the fluctuating Rashba field \( \xi \) (this is easily satisfied as \( \xi \sim 1 \) nm, while \( l \sim 10–100 \) nm in graphene), i.e. the electron spins experience indeed random spin–orbit coupling, the spin-flip scattering terms can be written as (appendix A) [32, 38]
\[ \partial_t \rho_{\mu k}(t) |_{scat}^{\prime} = -\frac{2\pi}{\hbar} \sum_{k'} C_{k-k'} \delta(\varepsilon_{\mu k} - \varepsilon_{\mu k'}) [\rho_{\mu k}(t) - V_{kk'} \rho_{\mu k'}(t) V_{k'k}]. \] (10)

Solving the KSBEs, one can obtain the spin relaxation properties from the time evolution of \( \rho_{\mu k}(t) \).

3. Analytical study of spin relaxation

In this section we analytically study the spin relaxation in graphene with the RRF. To realize this, we only take into account the spin-flip scattering as well as the strong elastic electron–impurity scattering. When the valley index is further omitted due to the degeneracy, the KSBEs are simplified as
\[ \partial_t \rho_k(t) = -\frac{i}{\hbar} [\mathbf{\Omega}_k \cdot \mathbf{\sigma}, \rho_k(t)] - \frac{2\pi}{\hbar} \sum_{k'} |U_{k-k'}|^2 I_{kk'} \delta(\varepsilon_k - \varepsilon_{k'}) [\rho_k(t) - V_{kk'} \rho_{k'}(t) V_{k'k}]. \] (11)
Here \( |U_{k-k'}|^2 \) is the effective electron–impurity scattering matrix element and \( I_{kk'} = \frac{1}{2} [1 + \cos(\theta_k - \theta_{k'})] \) is the form factor [31]. \( |U_{k-k'}|^2 = n_i^a |U_{k-k'}^a|^2 + n_i^s |U_{k-k'}^s|^2 \), with the first (second) term corresponding to the scattering of electrons from adatoms (impurities in the substrate). \( |U_{k-k'}^{a/s}|^2 \) is the electron–impurity Coulomb potential scattering matrix element [31, 34, 42]. \( n_i^a \) is the impurity density in the substrate. By defining the spin vector as \( \mathbf{S}_k(t) = \text{Tr}[\rho_k(t) \mathbf{\sigma}] \), one obtains the equation of \( \mathbf{S}_k(t) \) directly from equation (11) as
\[ \partial_t \mathbf{S}_k(t) = \frac{2\lambda_0}{\hbar} \mathbf{A}_k \cdot \mathbf{S}_k(t) - \frac{2\pi}{\hbar} \sum_{k'} |U_{k-k'}|^2 I_{kk'} \delta(\varepsilon_k - \varepsilon_{k'}) [\mathbf{S}_k(t) - \mathbf{S}_{k'}(t)] \]
\[ -\frac{2\pi}{\hbar} \sum_{k'} C_{k-k'} \delta(\varepsilon_k - \varepsilon_{k'}) [\mathbf{S}_k(t) - \mathbf{B}_{kk'} \cdot \mathbf{S}_k(t)]. \] (12)
Here
\[
A_k = \begin{pmatrix}
0 & 0 & \cos \theta_k \\
0 & 0 & \sin \theta_k \\
-\cos \theta_k & -\sin \theta_k & 0
\end{pmatrix}
\] (13)
and
\[
B_{kk'} = \begin{pmatrix}
-\cos(\theta_k + \theta_{k'}) & -\sin(\theta_k + \theta_{k'}) & 0 \\
-\sin(\theta_k + \theta_{k'}) & \cos(\theta_k + \theta_{k'}) & 0 \\
0 & 0 & -1
\end{pmatrix}.
\] (14)

By expanding \( \mathbf{S}(t) \) as \( \mathbf{S}(t) = \sum \mathbf{S}_l(t) e^{i\omega_l t} \) and retaining the lowest three orders of \( \mathbf{S}_l(t) \) (i.e. terms with \( l = 0, \pm 1 \)), one obtains a group of differential equations of \( \mathbf{S}_k^{\pm1,0}(t) \) (appendix B). With initial conditions, these equations can be solved and information on spin relaxation is obtained from \( \mathbf{S}_k(t) \). We label the spin relaxation rate along the \( x-\), \( y- \) or \( z- \)axis for states with momentum \( k \) as \( \Gamma_x \), \( \Gamma_y \) or \( \Gamma_z \), respectively. One has (appendix B)
\[
\Gamma_z = 2\Gamma_x = 2\Gamma_y = 2/\tau_{ks}^0 + 4\lambda_0^2\tau_{s}^1/\hbar^2,
\] (15)
where
\[
\frac{1}{\tau_{ks}^0} = \frac{k}{4\pi \hbar^2 v_f} \int_0^{2\pi} d\theta |U_{ql}|^2 \sin^2 \theta
\] (16)
and
\[
\frac{1}{\tau_{ks}^1} = \frac{k}{2\pi \hbar^2 v_f} \int_0^{2\pi} d\theta C_q.
\] (17)
with \( |U_{ql}|^2 \) and \( C_q \) depending only on \( |q| = 2k \sin \frac{\theta}{2} \). For a highly degenerate electron system in graphene, the spin relaxation is contributed by the spin-polarized electrons around the Fermi circle. Therefore, one can approximately obtain the spin relaxation rate of the whole electron system by replacing \( k \) with \( k_f \) in equation (15).

From equation (15), one notes that the spin relaxation rate (take \( \Gamma_z \) as an example) consists of two parts, \( \Gamma_{flip} = 2/\tau_{ks}^0 \), determined by the spin-flip scattering due to the RRF, and \( \Gamma_{DP} = 4\lambda_0^2\tau_{s}^1/\hbar^2 \), determined by the average Rashba field due to the DP mechanism. These two mechanisms contribute to spin relaxation independently. It is noted that \( \Gamma_{flip} \) obtained here is consistent with that given in [32] except that the correlation functions are different and \( \Gamma_{DP} \) is the one previously given in [34]. In the following we discuss the spin relaxation due to the two mechanisms respectively and compare their relative importance.

### 3.1. Spin relaxation caused by the spin-flip scattering due to the random Rashba field

By utilizing equation (3), one can obtain the spin relaxation rate along the \( z- \)axis due to the spin-flip scattering as
\[
\Gamma_{flip} = \frac{k_f}{\pi \hbar^2 v_f} \int_0^{2\pi} d\theta C_q
\]
\[
= \frac{8\pi^2 n_i^3 \delta^2 k_f}{\hbar^2 v_f} e^{-2\xi^2 k_f^2 I_0(2\xi^2 k_f^2)}
\]
\[
= \frac{8\pi^2 n_i^3 \delta^2 k_f^3}{\hbar^2 v_f} F(\xi k_f) = \frac{8\pi^2 n_i^3 \delta^2 k_f^3}{\hbar^2 v_f} G(\xi k_f).
\] (18)
In the limits $k_f \xi \ll 1$ and $k_f \xi \gg 1$,

$$\Gamma_{\text{flip}} \approx \frac{8\pi^2 n_e^{3/2}}{\hbar^2 v_f} \begin{cases} \frac{\xi^4 k_f}{(2\sqrt{\pi})} & (k_f \xi \ll 1) \\ \xi^3/(2\sqrt{\pi}) & (k_f \xi \gg 1). \end{cases} \tag{19}$$

In the above equations, $I_0(x)$ is the modified Bessel function, $F(x) = x e^{-2x^2} I_0(2x^2)$ and $G(x) = x^3 F(x)$.

We now focus on the various factors affecting $\Gamma_{\text{flip}}$. $\Gamma_{\text{flip}}$ is proportional to the adatom density $n_{e\text{a}}$, as expected. From the $x$ dependence of $F(x)$ and $G(x)$ as shown in figure 1, one can explore the dependence of $\Gamma_{\text{flip}}$ on $k_f$ and $\xi$, respectively. It is found that with an increase of $k_f$, $\Gamma_{\text{flip}}$ first increases almost linearly when $k_f \xi \leq 0.63$, then decreases mildly and eventually saturates $[F(\infty) = \frac{1}{\sqrt{\pi}}]$. As a result, with an increase of electron density $n_e$, $\Gamma_{\text{flip}}$ shows nonmonotonic behaviour with a peak located at $n_e^c = \frac{1}{\pi} (\frac{0.63}{\xi})^2$. When $n_e \ll n_e^c$, $\Gamma_{\text{flip}}$ is proportional to $n_e^{1/2}$, and when $n_e \gg n_e^c$, $\Gamma_{\text{flip}}$ becomes insensitive to $n_e$. These features are consistent with those presented in [32]. We give an estimation of $n_e^c$ here. When the correlation length $\xi$ is set to 1 nm, $n_e^c \approx 1.3 \times 10^{13}$ cm$^{-2}$, which is quite a high value compared to the experimental data. Usually, $n_e$ varies around $10^{12}$ cm$^{-2}$. Therefore, in order to observe the nonmonotonic behaviour of $\Gamma_{\text{flip}}$ with increasing $n_e$, $\xi$ is required to be around a relatively large value, e.g. 3.5 nm. When the electron density is fixed, $\Gamma_{\text{flip}}$ increases monotonically with increasing $\xi$, as indicated by the $x$ dependence of $G(x)$ in figure 1. The effect of the temperature $T$ on $\Gamma_{\text{flip}}$ can be inferred from the $k$ dependence of $\Gamma_{\text{flip}}$. For the highly degenerate electron system, $\Gamma_{\text{flip}}$ is insensitive to $T$. When the electron density is relatively low, with increasing $T$, $\Gamma_{\text{flip}}$ is expected to increase mildly as electrons tend to occupy the states with larger momentum. With typical values $\delta = 25$ meV$^2$, $n_{e\text{a}}^3 = 3 \times 10^{12}$ cm$^{-2}$, $n_e = 10^{12}$ cm$^{-2}$ and $\xi = 0.5$ nm, one has $k_f \xi \approx 0.09$ and $\Gamma_{\text{flip}}^{-1} \approx 670$ ps. If $\xi$ is made two times larger, i.e. 1 nm, $\Gamma_{\text{flip}}^{-1}$ becomes about 16 times smaller, as $\Gamma_{\text{flip}}$ is proportional to $\xi^4$ when $k_f \xi \ll 1$ (equation (19)).
3.2. Spin relaxation caused by the D’yakonov–Perel’ mechanism

In the analytical study, only elastic spin-conserving scattering is considered. With the other kinds of spin-conserving scattering included, the spin relaxation rate due to the DP mechanism should be modified to

\[
\Gamma_{\text{DP}} = 4\lambda_0^2 \tau_{\text{DP}}^* (k_t) / \hbar^2,
\]

where \(\tau_{\text{DP}}^* (k_t)\) is the effective momentum relaxation time limited by all the different kinds of scattering, including the electron–electron Coulomb scattering \([40]\). When \(\lambda_0\) is fixed, the electron density and temperature affect \(\Gamma_{\text{DP}}\) via \(\tau_{\text{DP}}^* (k_t)\). It has been shown previously \([31]\) that with an increase of \(n_e\) or a decrease of \(T\), the scattering strength decreases and \(\Gamma_{\text{DP}}\) increases. However, when the electron-impurity scattering is dominant, \(\Gamma_{\text{DP}}\) is insensitive to \(T\). The dependence of \(\Gamma_{\text{DP}}\) on \(n_e^a\) is not obvious. To facilitate the investigation, we write \(\tau_{\text{DP}}^{-1} (k_t) = \tau_{\text{DP}}^{-1} (k_t) + \tau_{\text{DP}}^{-1} (k_t)\), where \(\tau_{\text{DP}}^{-1} (k_t)\) is contributed by the electron-charged adatom scattering and \(\tau_{\text{DP}}^{-1} (k_t)\) by all the other kinds of scattering. From equation \((16)\), one has

\[
\tau_{\text{DP}}^{-1} (k_t) = \frac{n_e^a k_t}{4 \pi \hbar^2 v_f} \int_0^{2\pi} d\theta |U_q|^2 \sin^2 \theta = c_1 n_e^a
\]

with \(|q| = 2k_f \sin \theta / 2\). With \(\lambda_0 = \lambda_0^1 + 2\pi n_e^3 \bar{\delta} \xi^2 = \lambda_0^1 + c_2 n_e^a\), \(\Gamma_{\text{DP}}\) can be written as

\[
\Gamma_{\text{DP}} = \frac{4}{\hbar^2} \tau_{\text{DP}}^{-1} (k_t) + c_1 n_e^a
\]

which indicates a complex dependence on \(n_e^a\). When \(c_2 = 0\) and \(\lambda_0^1 \neq 0\), \(c_2 > 0\) and \(0 \leq n_e^a \leq c_2^{-1} \lambda_0^1 - 2c_1^{-1} \tau_{\text{DP}}^{-1} (k_t)\) or \(c_2 < 0\) and \(0 \leq n_e^a \leq -c_2^{-1} \lambda_0^1\), \(\Gamma_{\text{DP}}\) decreases with increasing \(n_e^a\), exhibiting a DP-like behaviour. However, most interestingly, when \(c_2 > 0\) and \(n_e^a \geq c_2^{-1} \lambda_0^1 - 2c_1^{-1} \tau_{\text{DP}}^{-1} (k_t)\) or \(c_2 < 0\) and \(n_e^a \geq -c_2^{-1} \lambda_0^1\), \(\Gamma_{\text{DP}}\) increases with increasing \(n_e^a\), exhibiting EY-like behaviour. In particular, here in the limit with \(n_e^a\) being large enough, \(\Gamma_{\text{DP}} \propto n_e^a\) approximately. With typical values in the presence of adatoms \([34]\), \(\lambda_0 = 0.2\text{ meV}\) and \(\tau_{\text{DP}}^* (k_t) = 0.1\text{ ps}\), \(\Gamma_{\text{DP}}^{-1}\) is calculated to be about 100 ps.

3.3. Comparison of relaxations caused by the spin-flip scattering due to the random Rashba field and the D’yakonov–Perel’ mechanism

In this subsection we discuss the relative importance of the mechanism of spin-flip scattering due to the RRF and the DP mechanism in the regime with \(k_t \xi \ll 1\), which is typically realized in graphene. Under this condition, from equations \((19)\) and \((22)\), one obtains \(\Gamma_{\text{DP}} / \Gamma_{\text{flip}} \approx 10 \bar{\delta}^2 / \bar{\delta}^2\) when \(\lambda_0^1 = \tau_{\text{flip}}^{-1} (k_t) = 0\). For the case with single-sided adatoms, \(\bar{\delta}^2 = \bar{\delta}^2\); therefore \(\Gamma_{\text{DP}} / \Gamma_{\text{flip}} \approx 10\) and \(\Gamma_{\text{flip}}\) can be neglected. However, for the case with double-sided adatoms, \(\Gamma_{\text{flip}}\) can be comparable to or can even surpass \(\Gamma_{\text{DP}}\) as \(\bar{\delta}\) may be as small as zero. In reality \(\tau_{\text{flip}}^{-1} (k_t) \neq 0\) and \(\Gamma_{\text{DP}}\) decreases with increasing \(\tau_{\text{flip}}^{-1} (k_t)\). When the substrate also contributes to the Rashba field as \(\lambda_0^1\), \(\Gamma_{\text{DP}}\) can be either enhanced (e.g. when \(\lambda_0^1 \bar{\delta} > 0\) or suppressed. As a consequence, for the configuration with single-sided adatoms, when the contribution from the substrate to the average Rashba field does not compensate that from the adatoms (e.g. when \(\lambda_0^1 \bar{\delta} \geq 0\)) and the scattering other than the electron–adatom type is not extraordinarily strong (i.e. \(\tau_{\text{flip}}^{-1}\) is not unusually large), the spin relaxation caused by the spin-flip scattering due to the RRF can be neglected. In such
In a case, the spin relaxation is limited by the DP mechanism with the adatoms contributing to the average Rashba field. This is just how the effect of adatoms was incorporated into our previous investigation \[34\]. For other cases, whether the spin-flip scattering due to the RRF is important or not when compared to the DP mechanism is condition dependent. Undoubtedly, when the average Rashba field approaches zero, the spin-flip scattering due to the RRF tends to be important. In the work of Dugaev et al \[32\], the average Rashba field induced by ripples is zero and the spin relaxation is solely determined by the spin-flip scattering due to the RRF. However, the spin relaxation time calculated in their model is of the order of 10 ns, two orders of magnitude larger than the experimental values.

4. Numerical results

The KSBEs need to be solved numerically in order to take full account of all the different kinds of scattering. The initial conditions are set as

\[
\rho_{\mu k}(0) = \frac{f_{k}^{0} + f_{-k}^{0}}{2} + \frac{f_{k}^{0} - f_{-k}^{0}}{2} \hat{n} \cdot \sigma, \tag{23}
\]

\[
\sum_{\mu k} \text{Tr}[\rho_{\mu k}(0) \hat{n} \cdot \sigma] = n_e P(0), \tag{24}
\]

\[
\sum_{\mu k} \text{Tr}[\rho_{\mu k}(0)] = n_e. \tag{25}
\]

At time \(t = 0\), the electrons are polarized along \(\hat{n}\) with the density and spin polarization being \(n_e\) and \(P(0)\), respectively. \(f_{k}^{0}\) is the Fermi distribution function of electrons with spins parallel/antiparallel to \(\hat{n}\), where the chemical potential is determined by equations \((24)–(25)\). By solving the KSBEs, one can obtain the time evolution of spin polarization along \(\hat{n}\) as \(P(t) = \frac{1}{n_e} \sum_{\mu k} \text{Tr}[\rho_{\mu k}(t) \hat{n} \cdot \sigma]\) and hence the spin relaxation time \(\tau_s\). In the calculation, we set \(P(0)\) to be as small as 0.05 and \(\hat{n}\) in the graphene plane, such as, along the \(y\)-axis, in order to compare with experiments.

4.1. Adatom density dependence of spin relaxation

In this section, we study the adatom density dependence of spin relaxation based on the single-sided adatom model. In figure 2, the in-plane spin relaxation rate against the adatom density (at the top of the frame) or the inverse of charge diffusion coefficient (at the bottom of the frame) is shown. The temperature is \(T = 300\) K, the electron density is \(n_e = 10^{12}\) cm\(^{-2}\), the density of impurities in the substrate is \(n_s = 0.2 \times 10^{12}\) cm\(^{-2}\) and the parameters for the single-sided adatom model are \(\delta = 5\) meV and \(\xi = 0.5\) nm. In the figure, the spin relaxation rates with different values of \(\lambda_i^0\) are plotted by the curves. The nearby data points of each curve are calculated with the spin-flip scattering being removed. The small discrepancy between each curve and corresponding data points indicates that the DP mechanism dominates the spin relaxation. It is noted that in a large parameter regime of the background Rashba field \(\lambda_i^0\), the curves show obvious EY-like behaviour, i.e. the spin relaxation rate is proportional to the momentum relaxation rate. When \(\lambda_i^0\) is large enough (larger than \(2c_2c_1^{-1} \tau_{\text{p}}^{-1}(k_f) \approx 0.05\) meV from the discussion in section 3.2), the spin relaxation rate decreases with increasing adatom density at low doping density of adatoms, showing DP-like behaviour.
We further apply the single-sided adatom model to reinvestigate the experiment of Pi et al from Riverside [24], which shows obvious DP-like behaviour. At $T = 18$ K, with the increasing density of adatoms (Au atoms) from surface deposition (although Au atoms also donate electrons to graphene, the electron density is fixed at $2.9 \times 10^{12}$ cm$^{-2}$ by adjusting the gate voltage [24]), the diffusion coefficient decreases while the spin relaxation time increases, as indicated by the crosses with error bars in figure 3. By fitting the experimental data without adatoms (before Au deposition), we obtain $\lambda^i_0 = 0.153$ meV and $n^i_s = 2.1 \times 10^{12}$ cm$^{-2}$ [34]. During Au deposition, a group of parameters, $\delta = 2.03$ meV and $\xi = 0.5$ nm, can reproduce the experimental data except when the adatom density $n^a_i$ is larger than $2 \times 10^{12}$ cm$^{-2}$ (the solid curve). By assuming that $\xi$ decreases with an increase of $n^a_i$ when $n^i_s$ is large enough (the inset of figure 3), the recalculation can cover the experimental data in the region with large $n^a_i$ (the dashed curve). Similar to figure 2, the dots nearby the solid curve are calculated with the spin-flip scattering being removed (with $\xi$ being fixed at 0.5 nm). The small discrepancy between the solid curve and dots indeed indicates that the spin-flip scattering due to the RRF is not important.

**Figure 2.** The dependence of in-plane spin relaxation rate on the adatom density (at the top of the frame) or the inverse of the charge diffusion coefficient (at the bottom of the frame). The temperature is $T = 300$ K, the electron density is $n_e = 10^{12}$ cm$^{-2}$, the density of impurities in the substrate is $n^i_s = 0.2 \times 10^{12}$ cm$^{-2}$ and the parameters for the single-sided adatom model are $\delta = 5$ meV and $\xi = 0.5$ nm. The curves are calculated with different $\lambda^i_0$. For each curve, its nearby data points are calculated with the same parameters but without the spin-flip scattering.

4.2. Electron density dependence of spin relaxation

The electron density dependence of spin relaxation is studied by fitting the experiment of Józsa et al from Groningen [23], which shows EY-like behaviour. At room temperature, with an increase of electron density from 0.16 to $2.81 \times 10^{12}$ cm$^{-2}$ (adjusted by the gate voltage),
Figure 3. The dependence of in-plane spin relaxation rate on the adatom density (at the top of the frame) or the inverse of the charge diffusion coefficient (at the bottom of the frame). The temperature is $T = 18$ K, the electron density is $n_e = 2.9 \times 10^{12}$ cm$^{-2}$ and the density of impurities in the substrate is $n_i = 2.1 \times 10^{12}$ cm$^{-2}$. The crosses with error bars are the experimental data from Pi et al [24]. The solid curve stands for the fitting data via the single-sided adatom model with $\lambda_0 = 0.153$ meV, $\delta = 2.03$ meV and $\xi = 0.5$ nm. The nearby dots of the solid curve are calculated with the spin-flip scattering terms removed. The dashed curve is calculated with the same parameters as that of the solid curve except that $\xi$ decreases with $n_a$ when $n_a > 2 \times 10^{12}$ cm$^{-2}$, as shown in the inset.

both the charge diffusion coefficient and the spin relaxation time increase, with the latter being proportional to the former (squares in figure 4). It should be noted that this EY-like behaviour cannot be explained by the nearly linear curves shown in figure 2, as the linearity there is due to an increase of adatom density when the electron density is fixed. In fact, according to section 3, with the increase of $n_e$ as well as the accompanying increase of $D$ (and hence the increase of $\tau_p^*(k_f)$), both $\Gamma_{\text{flip}}$ and $\Gamma_{\text{DP}}$ should increase when the parameters for the adatom model are fixed (see equations (19) and (22)). However, as will be shown in the following, with the assumption that $\xi$ decreases with increasing $n_e$, both the single-sided and double-sided adatom models (hence both the DP mechanism and the mechanism of the spin-flip scattering due to the RRF) are able to fit the experimental data.

In figure 4, we present the fitting to the experimental data of Józsa et al via the single-sided adatom model where the DP mechanism is dominant. The calculated spin relaxation time is plotted by the open circles in figure 4. The fitting parameters are chosen as $\lambda_0 = 0.127$ meV, $n_i = 0.5 \times 10^{12}$ cm$^{-2}$ and $\delta = 4$ meV here. In order to reproduce the electron density dependence of diffusion coefficient, the impurity density in the substrate $n_i$ has to increase with increasing $n_e$ possibly due to the increased ionization (otherwise if $n_i$ is fixed, $D$ will increase with increasing $n_e$ much more quickly), as shown by the dots in the inset. Meanwhile, with
Figure 4. Fit to the experimental data (the dependence of spin relaxation time $\tau_s$ on the charge diffusion coefficient $D$ with an increase of $n_e$) of Józsa et al [23] via the single-sided adatom model. The squares stand for the experimental data and the open circles are from our calculation. The inset shows the dependences of $\xi$ (with the scale on the right-hand side of the frame) and $n_i^s$ on $D$ when $n_e$ is increased. For comparison, the dashed curve is calculated with $\xi$ being fixed at 1.05 nm. $n_i^s = 0.5 \times 10^{12}$ cm$^{-2}$, $\lambda_0^i = 0.127$ meV and $\delta = 4$ meV.

the increase of $n_e$, $\xi$ should decrease as shown by the triangles in the inset (the scale is on the right-hand side of the frame) to account for the increase of $\tau_s$. Otherwise if $\xi$ is fixed, $\tau_s$ will decrease with increasing $D$ mainly due to the increase of $\Gamma_{DP}$, as indicated by the dashed curve in the figure. In figure 5, we also present a feasible fitting by the double-sided adatom model. The squares stand for the experimental data and the open circles are from calculation. In our computation, $\lambda_0^i = 0$, $n_i^s = 0.5 \times 10^{12}$ cm$^{-2}$ and $\delta_1 = -\delta_2 = 5$ meV. The inset shows the dependences of $\xi$ (open triangles with the scale on the right-hand side of the frame) and $n_i^s$ (solid circles) on $D$ when $n_e$ is increased. In this fitting, only the spin-flip scattering due to the RRF plays a role in spin relaxation.

4.3. Temperature dependence of spin relaxation

We investigate the temperature dependence of spin relaxation in graphene in this section. Although the spin relaxation time determined by the DP mechanism increases with growing $T$ as pointed out in section 3, this dependence becomes very weak when the electron-impurity scattering is dominant (which is satisfied in graphene on SiO$_2$ substrate), as revealed by figure 1 of [31] (note that the mobility there is even one order of magnitude larger than the ones in this investigation). The spin relaxation time determined by the spin-flip scattering due to the RRF is also insensitive to $T$, as shown in figure 6. Therefore, when other parameters are fixed, the spin relaxation in graphene is expected to depend on temperature weakly.

It is quite interesting that a decrease of $\tau_s$ with $T$ was observed by the Riverside group very recently [26]. Moreover, when $T$ is fixed, with an increase of $n_e$ (adjusted by the gate
Figure 5. Fit to the experimental data (the dependence of spin relaxation time $\tau_s$ on the charge diffusion coefficient $D$ with an increase of $n_e$) of Józsa et al [23] via the double-sided adatom model. The squares stand for the experimental data and the open circles are from our calculation. The inset shows the dependences of $\xi$ (with the scale on the right-hand side of the frame) and $n_i$ on $D$ when $n_e$ is increased. $n_i^0 = 0.5 \times 10^{12} \text{ cm}^{-2}$, $\lambda^0_i = 0$ and $\delta_1 = -\delta_2 = 5 \text{ meV}$.

Figure 6. Temperature dependence of spin relaxation time due to the spin-flip scattering. The parameters are $n_e = 1.5 \times 10^{12} \text{ cm}^{-2}$, $n_i^0 = 0.5 \times 10^{12} \text{ cm}^{-2}$ and $\delta^2 = 4 \text{ meV}^2$. Solid curve: $\xi = 0.5 \text{ nm}$; and dotted curve: $\xi = 0.8 \text{ nm}$.

voltage), both $D$ and $\tau_s$ increase, similar to the observations of Józsa et al [23]. The decrease of $\tau_s$ with increasing $T$ may be due to the increase of the correlation length $\xi$ of the RRF with an increase of $T$, with either the DP mechanism or the spin-flip scattering due to the RRF being dominant. The linear scaling between $\tau_s$ and $D$ with the variation of electron density.
Figure 7. Fit to the experimental data of Han and Kawakami [26] with the single-sided adatom model. (a) and (b) The temperature dependence of $\tau_s$ and $D$. The squares (open circles) are the experimental data with $V_{\text{CNP}} = 20$ V (60 V), and the solid (dashed) curve is from fitting with $V_{\text{CNP}} = 20$ V (60 V). (c) The temperature dependences of $\xi$ and $n_i^s$ (on the right-hand side of the frame). The solid and dotted curves are for the case with $V_{\text{CNP}} = 20$ V, and the dashed and chain curves are for the case with $V_{\text{CNP}} = 60$ V.

As a feasible way, we fit the temperature dependence of spin relaxation based on the single-sided adatom model by assuming $\xi$ to increase with $T$. One possible fitting is shown in figure 7. Experimentally, when the gate voltage $V_{\text{CNP}} = 20$ V (60 V), the electron density $n_e = 1.47 \times 10^{12}$ cm$^{-2}$ (4.42 $\times 10^{12}$ cm$^{-2}$). In figures 7(a) and (b), the squares (open circles) are the experimental data of spin relaxation time and diffusion coefficient with $V_{\text{CNP}} = 20$ V (60 V), respectively, and the solid (dashed) curves are from our calculation with $V_{\text{CNP}} = 20$ V (60 V). The fitting parameters are $n_i^e = 0.5 \times 10^{12}$ cm$^{-2}$, $\lambda_{i0} = 0.052$ meV and $\delta = 2$ meV.

The electron density is obtained from the arXiv version of [26] (arXiv:1012.3435).
Figure 8. Fit to the experimental data (the dependence of spin relaxation time $\tau_s$ on the charge diffusion coefficient $D$ with an increase of $n_e$) of Han and Kawakami [26] via the single-sided adatom model. The solid squares stand for the experimental data and the open circles are from our calculation. The inset shows the dependences of $\xi$ (with the scale on the right-hand side of the frame) and $n_i^s$ on $D$ when $n_e$ is increased. The open squares are the experimental data for holes or near the charge neutrality point, which are not considered in our fitting. $n_a^i = 0.5 \times 10^{12} \text{ cm}^{-2}$, $\lambda_0^i = 0.052 \text{ meV}$ and $\delta = 2 \text{ meV}$.

variation of $\xi$ with $T$ is shown in figure 7(c), where the solid (dashed) curve is for $V_{\text{CNP}} = 20 \text{ V}$ ($60 \text{ V}$). The variation of $n_i^s$ with $T$ is also shown in figure 7(c) with the scale on the right-hand side of the frame, where the dotted (chain) curve is for $V_{\text{CNP}} = 20 \text{ V}$ ($60 \text{ V}$). In this fitting, the DP mechanism is dominant and the spin-flip scattering due to the RRF can be neglected. In fact, the calculation with similar parameters in figure 6 has indicated that the spin relaxation time caused by the spin-flip scattering due to the RRF is very long. With the same parameters, we further fit the dependence of spin relaxation time on diffusion coefficient at 100 K in figure 8. Consistent with the fittings in the previous section, the correlation length of the RRF is also found to decrease with increasing electron density. It is noted that the open squares in the figure are the data measured for holes or near the charge neutrality point [26] and hence are not considered in our fitting.

4.4. A nonmonotonic dependence of $\tau_s$ on $D$

In the experiments of both the Riverside [26] and Groningen [23] groups, the spin relaxation time $\tau_s$ is observed to depend monotonically on the diffusion coefficient $D$. However, very recently, a nonmonotonic dependence of $\tau_s$ on $D$ with an increase of carrier density at $T = 4.2 \text{ K}$ was reported by Jo et al [27]. Although this phenomenon is reported at the hole band, we can treat it at the electron band by our model due to the electron–hole symmetry of band structure in graphene. In figure 9, the experimental data are fitted by the single-sided adatom model with
Figure 9. Fit to the experimental data (the dependence of the spin relaxation time $\tau_s$ and charge diffusion coefficient $D$ on electron density $n_e$) of Jo et al [27] via the single-sided adatom model. The closed squares (solid triangles) are the experimental (fitting) data of the spin relaxation time $\tau_s$ and the open squares (solid circles) are the experimental (fitting) data of the diffusion coefficient $D$ (with the scale on the right-hand side of the frame). The inset shows the density dependence of $\xi$ (open triangles with the scale on the right-hand side of the frame) and $n_{si}$. $n_{si} = 0.5 \times 10^{12} \text{ cm}^{-2}$, $\delta = 6 \text{ meV}$.

$n_{si} = 0.5 \times 10^{12} \text{ cm}^{-2}$, $\lambda_0 = 0.18 \text{ meV}$ and $\delta = 6 \text{ meV}$. The closed squares (solid triangles) are the experimental (fitting) data of the spin relaxation time $\tau_s$ and the open squares (solid circles) are the experimental (fitting) data of the diffusion coefficient $D$ (with the scale on the right-hand side of the frame). The inset shows the density dependence of $\xi$ (open triangles with the scale on the right-hand side of the frame) and $n_{si}$ (solid circles). Due to the slower decrease of $\xi$ and the faster increase of $n_{si}$ with increasing $n_e$, it is possible for $\tau_s$ to increase with $n_e$ when the latter is large enough.

4.5. Possible factors affecting the correlation length of the random Rashba field

In our fittings to the experiments, the variation of the correlation length $\xi$ of the RRF plays an essential role. $\xi$ is found to decrease with increasing electron density and to increase with increasing temperature. $\xi$ is also found to decrease with an increase of adatom density when the latter is high enough. It is indeed quite possible that $\xi$ is affected by these factors. For example, the correlation length might be shortened by the screening of carriers, which is more effective when the carrier density is high. It is also possible that with increasing temperature the adatoms tend to form clusters and enhance the inhomogeneity [43]. Besides, the puddle size, which measures the correlation length of the short-range random potential in graphene, decreases with increasing density of the charged impurities [47]. A similar feature is also expected in the present study, i.e. $\xi$ decreases with increasing adatom density when the latter is large enough.
5. Discussion and conclusion

5.1. Discussion of the possible dominant spin relaxation mechanism

We now summarize our numerical fittings to the experiments and discuss the possible dominant spin relaxation mechanism. Fitting the DP-like behaviour with an increase of adatom density observed by the Riverside group [24], we find that the DP mechanism is the dominant one and that only with it can the experimental phenomenon be understood. It is noted that the correlation length $\xi$ of the RRF is supposed to be constant at the low density regime of adatoms, and when $\xi$ is fixed, $\Gamma_{\text{flip}}$ always increases with adatom density. Therefore, other kinds of attempts with the spin-flip scattering due to the RRF being dominant fail to reproduce the experimental phenomenon and can be ruled out. However, the EY-like behaviour with an increase of electron density observed by the Groningen group [23] can be fitted by our model with either the DP mechanism or the spin-flip scattering due to the RRF being dominant when the decrease of $\xi$ with increasing electron density is considered. Nevertheless, the fact that the Riverside group has also observed similar EY-like behaviour in their samples very recently [26], in combination with the observation of the adatom density dependence of the spin relaxation [24], suggests that the DP mechanism is dominant, but is exhibiting EY-like properties with increasing electron density. The similar experimental phenomena on the electron density dependence of the spin relaxation from the Groningen [23] and Riverside [26] groups further suggest that the DP mechanism also dominates the spin relaxation in the experiment of the Groningen group [23]. Consequently, with the DP mechanism being dominant in graphene, the RRF leads to spin relaxation which exhibits either DP- or EY-like properties in the experiments.

5.2. Conclusion

In conclusion, we have studied electron spin relaxation in graphene with random Rashba field using the kinetic spin Bloch equations, with the aim of understanding the main spin relaxation mechanism in graphene in the present experiments. Different from the case previously studied by Zhou and Wu where no adatoms are considered and the mobility is relatively high [1, 31], the electron mobility investigated in the present work is at least one order of magnitude smaller due to the extrinsic factors caused by the ferromagnetic electrodes used in the spin relaxation measurements [20–23, 26, 27], e.g. the adatoms. We set up a random Rashba model to incorporate the contribution of both the adatoms and the substrate. In this model, the charged adatoms, on the one hand, enhance the Rashba spin–orbit coupling locally and, on the other hand, serve as Coulomb potential scatterers.

Based on the random Rashba model, an analytical study on spin relaxation in graphene is conducted. The average of the random Rashba field leads to spin relaxation limited by the DP mechanism, which is absent in the work of Dugaev et al [32], while the randomness of the random Rashba field causes spin relaxation by spin-flip scattering, serving as an EY-like mechanism. With an increase of adatom density, the spin relaxation caused by the spin-flip scattering due to the random Rashba field always shows EY-like behaviour, whereas the DP mechanism can exhibit either EY- or DP-like one. When all the other parameters are fixed, with an increase of electron density the spin relaxation rates due to both mechanisms increase. Nevertheless, the spin relaxation rate determined by the spin-flip scattering due to the random Rashba field is insensitive to temperature, whereas that determined by the DP mechanism becomes insensitive to temperature when the electron-impurity scattering is dominant. However,
both mechanisms are sensitive to the correlation length of the random Rashba field, which may be affected by environmental factors such as electron density and temperature.

We further perform numerical calculations and fit the experiments of the Riverside [24, 26] and Groningen [23] groups, which show either DP or EY-like properties. Fitting and comparing these experiments, we suggest that the DP mechanism dominates the spin relaxation in graphene. With the DP mechanism being dominant, the random Rashba field leads to spin relaxation which exhibits either DP or EY-like properties. Besides, the nonmonotonic dependence of $\tau_s$ on $D$ very recently reported by Jo et al [27] is also fitted by our model, with the DP mechanism being dominant.

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Appendix A. Spin-flip scattering terms

The spin-flip scattering terms are [38]

$$\partial_t \rho_{\mu k}(t)|_{\text{scat}} = -\frac{\pi}{\hbar} \sum_{k' \neq k} |\tilde{\lambda}_{k'-k}|^2 \delta(\epsilon_{\mu k} - \epsilon_{\mu k'}) [\rho_{\mu k}(t) V_{kk'} V_{k'k} + V_{kk} V_{k'k} \rho_{\mu k}(t)]$$

$$-2V_{kk} \rho_{\mu k'}(t) V_{k'k} \]$$

\[= -\frac{2\pi}{\hbar} \sum_{k'} |\tilde{\lambda}_{k-k'}|^2 \delta(\epsilon_{\mu k} - \epsilon_{\mu k'}) [\rho_{\mu k}(t) - V_{kk} \rho_{\mu k'}(t) V_{k'k}], \tag{A.1} \]

where

$$\tilde{\lambda}_q = \int [\lambda(r) - \lambda_0] e^{-iq \cdot r} \, dr \tag{A.3}$$

satisfying $\tilde{\lambda}_q = 0$ and $\tilde{\lambda}_q \neq 0 = \lambda_q$. When the mean free path $l$ is much larger than the correlation length $\xi$ of the fluctuating Rashba field, $|\tilde{\lambda}_q|^2$ can be approximated by its statistical average as follows:

$$|\tilde{\lambda}_q|^2 \approx \iint dr \, dr' \langle (\lambda(r) - \lambda_0)(\lambda(r') - \lambda_0) \rangle e^{-iq \cdot (r-r')} = \iint dr \, dr' C(r-r') e^{-iq \cdot (r-r')} \approx C_q. \tag{A.4}$$

Equations (A.2) and (A.4) lead to equation (10).

Appendix B. Analytical study of spin relaxation in graphene

We present the analytical study of spin relaxation in graphene in detail. By expanding $S_k(t)$ as $S_k(t) = \sum_{l} S_{k}^{l}(t) e^{il\theta_k}$, one obtains from equation (12) the following equations:

$$\partial_t S_{kx}^{l}(t) = \frac{\lambda_0}{\hbar} \sum_{l_0=\pm 1} S_{kz}^{l+l_0}(t) - \left( \frac{1}{\tau_k^{+}} + \frac{1}{\tau_{ks}^{-}} \right) S_k^{l}(t) - \sum_{l_0=\pm 1} \frac{S_{kx}^{l+2l_0}(t) + i\frac{\lambda_0}{\hbar} S_{kx}^{l}(t) + i\frac{\lambda_0}{\hbar} S_{kx}^{l+2l_0}(t)}{2\tau_{kx}^{l_0}}, \tag{B.1}$$

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\[ \frac{\partial}{\partial t} S_{kx}^l(t) = \frac{\lambda_0}{\hbar} \sum_{l_0=\pm1} i_0 S_{kx}^{l+l_0}(t) - \left( \frac{1}{\tau_k} + \frac{1}{\tau_{ks}} \right) S_{kx}^l(t) - \sum_{l_0=\pm1} \frac{i_0 S_{kx}^{l+l_2l_0}(t) - S_{kx}^{l+2l_0}(t)}{2\tau_{kx}^l}, \]  

(B.2)

\[ \frac{\partial}{\partial t} S_{ky}^l(t) = -\frac{\lambda_0}{\hbar} \bar{\hbar} \sum_{l_0=\pm1} [S_{kx}^{l+l_0}(t) + i_0 S_{ky}^{l+l_0}(t)] - \left( \frac{1}{\tau_k} + \frac{2}{\tau_{ks}} \right) S_{ky}^l(t), \]  

(B.3)

in which

\[ \frac{1}{\tau_k^l} = \frac{k}{4\pi \bar{\hbar}^2 v_f} \int_0^{2\pi} d\theta |U_q|^2 (1 + \cos \theta)(1 - \cos l\theta) \]  

(B.4)

and

\[ \frac{1}{\tau_{ks}^l} = \frac{k}{2\pi \bar{\hbar}^2 v_f} \int_0^{2\pi} d\theta C_q \cos l\theta. \]  

(B.5)

Here \(|U_q|^2\) and \(C_q\) depend only on \(|q| = 2k \sin \frac{\theta}{2}\). It is noted that \(\frac{1}{\tau_k^l} = \frac{1}{\tau_k}\) and \(\frac{1}{\tau_{ks}^l} = \frac{1}{\tau_{ks}}\). It is also noted that \(\frac{1}{\tau_{ks}^l} = 0\) and \(\tau_k^l\) is in fact the momentum relaxation time \(\tau_k(q, k)\) limited by the electron-impurity scattering.

By retaining the lowest three orders of \(S_k^l(t)\) (i.e. terms with \(l = 0, \pm 1\)) in equations (B.1)–(B.3), one obtains

\[ \begin{pmatrix} \partial_t & -F & P & Q \\ -P^T & G & P & Q \\ Q^T & -P^T & F \end{pmatrix} \begin{pmatrix} S_k^0 \\ S_k^1 \\ S_k^{-1} \end{pmatrix} = 0, \]  

(B.6)

where

\[ G = -\frac{1}{2\tau_{ks}^0} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix}, \]  

(B.7)

\[ F = G - \frac{1}{\tau_k^0}, \]  

(B.8)

\[ P = \frac{\lambda_0}{\hbar} \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & -i \\ -1 & i & 0 \end{pmatrix}, \]  

(B.9)

\[ Q = \frac{1}{2\tau_{ks}^0} \begin{pmatrix} -1 & i & 0 \\ i & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \]  

(B.10)

As the spin flipping rate \(1/\tau_{ks}^0\) is much lower than the momentum relaxation rate \(1/\tau_k^1\) (in graphene \(1/\tau_k^1\) is usually of the order of \(10 \text{ ps}^{-1}\); even if \(1/\tau_{ks}^0\) reaches the experimental value \(\sim 0.01 \text{ ps}^{-1}\), \(\tau_k^1/\tau_{ks}^0\) is still as small as \(10^{-3}\) and \(S_k^\pm\) are smaller terms compared to \(S_k^0\) in the strong scattering limit, we approximate \(F\) and \(Q\) as \(F \approx -\frac{1}{\tau_k^1}\) and \(Q \approx 0\). With the initial
conditions, e.g. $S_l^0(0) = \delta_{l0}(0, 0, S_{kz}^0(0))^T$ ($l = 0, \pm 1$), equation (B.6) can be solved as (those of $S_{kz}^{\pm1}(t)$ are not of interest to us and are not shown here)

$$S_k^0(t) = \frac{1}{2} \left[ \left( 1 + \frac{1}{\sqrt{1 - c_z^2}} \right) e^{-\Gamma_1^- t} + \left( 1 - \frac{1}{\sqrt{1 - c_z^2}} \right) e^{-\Gamma_1^+ t} \right] (0, 0, S_{kz}^0(0))^T,$$

(B.11)

where

$$c_z = \frac{4\lambda_0}{\hbar} \frac{1}{1/\tau_k^1 - 1/\tau_{ks}^1}$$

(B.12)

and

$$\Gamma_{\pm} = \frac{1}{\tau_{kz}^0} \pm \frac{1}{2 \tau_k^1} \left( \frac{1}{\tau_{ks}^0} - \frac{1}{2 \tau_k^1} \right) \sqrt{1 - c_z^2}.$$  

(B.13)

In the strong scattering limit with $c_z \ll 1$, $S_{kz}^0(t) \approx S_{kz}^0(0)e^{-\Gamma_1^- t}$ where $\Gamma_{\pm} \approx 2/\tau_{kz}^0 + 4\lambda_0^2 \tau_k^1/\hbar^2$. Consequently, one obtains

$$S_k^0(t) = e^{-\Gamma_1^- t} (0, 0, S_{kz}^0(0))^T$$

(B.14)

with

$$\Gamma_\pm = 2/\tau_{kz}^0 + 4\lambda_0^2 \tau_k^1/\hbar^2.$$  

(B.15)

Similarly, with $S_l^0(0) = \delta_{l0}(S_{kz}^0(0), 0, 0)^T$ or $S_l^0(0) = \delta_{l0}(0, S_{ky}^0(0), 0)^T$, $S_k^0(t)$ is solved to be

$$S_k^0(t) = e^{-\Gamma_1^- t} (S_{kz}^0(0), 0, 0)^T$$

(B.16)

and

$$S_k^0(t) = e^{-\Gamma_1^- t} (0, S_{ky}^0(0), 0)^T,$$

(B.17)

respectively, with $\Gamma_x = \Gamma_y = \Gamma_\pm/2$.

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