Intrinsic electron spin relaxation due to the D’yakonov-Perel’ Mechanism in monolayer MoS\textsubscript{2}

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Intrinsic electron spin relaxation due to the D’yakonov-Perel’ mechanism is studied in monolayer Molybdenum Disulphide with the electron-impurity, electron-electron Coulomb, (both the intra- and inter-valley) electron-phonon scatterings explicitly included. We find that only the intervalley electron-phonon scattering contributes to the intrinsic in-plane spin relaxation whereas the contributions of other scatterings are absent. This originates from the unique momentum-independent intrinsic spin-orbit coupling, which only serves as a Zeeman-like term with opposite effective magnetic fields perpendicular to the monolayer Molybdenum Disulphide plane in the two valleys. The Zeeman-like term opens an intervalley spin relaxation channel together with the intervalley electron-phonon scattering. The intervalley electron-phonon scattering is always in the weak scattering limit, which makes the in-plane spin relaxation time proportional to the intervalley momentum scattering time. This leads to a rapid decrease of the in-plane spin relaxation time with increasing temperature due to the enhancement of the intervalley scattering. In addition, we predict a peak in the electron-density dependence of the in-plane spin relaxation time. This peak results from the crossover between the degenerate and nondegenerate limits. We also find that the in-plane spin relaxation time decreases rapidly with the increase of the initial spin polarization at low temperature, which is the same as the situation in bilayer graphene but opposite to the cases of both semiconductors and single-layer graphene.

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I. INTRODUCTION

Transition-metal dichalcogenides, similar to graphite, are layered materials with weak interlayer van der Waals interaction. These materials can be exfoliated to single to few-layer samples as the fabrication of graphene.\textsuperscript{1–5} Among these samples, monolayer Molybdenum Disulphide (MoS\textsubscript{2}) has received much attention due to its distinctive properties.\textsuperscript{5–11} It has a direct gap at the inequivalent K and K’ points,\textsuperscript{7,11,22,23,32,34} which makes it attractive as a two-dimensional channel material in field-effect transistors. Very recently, the field-effect transistor devices based on monolayer MoS\textsubscript{2} have been realized in the experiments with high on-off ratio.\textsuperscript{5,28} In addition, space inversion symmetry is broken in monolayer MoS\textsubscript{2} since it is a two-dimensional hexagonal lattice consisted of two different sublattices i.e., Mo and S atoms. The absence of the space inversion symmetry results in a valley-dependent optical selection rule for interband transitions, which allows the realization of the valley polarization by optical pumping with circularly polarized light.\textsuperscript{6,11,14–16,18,20,24} Space inversion symmetry breaking can also induce spin splitting of both the conduction and valence bands.\textsuperscript{11,20,23,27,31,35,38} This spin splitting is essential for spin physics and spintronic applications. All these intriguing features make monolayer MoS\textsubscript{2} of particular interest.

As spin relaxation is crucial for possible realistic spintronic applications, a thorough understanding of the spin relaxation time (SRT) in monolayer MoS\textsubscript{2} is essential. Very recently, Ochoa and Roldán\textsuperscript{37} investigated the intravalley spin-orbit mediated spin relaxation in monolayer MoS\textsubscript{2}. For electrons, they calculated the out-of-plane spin relaxation due to the extrinsic Rashba spin-orbit coupling (SOC) induced by an external out-of-plane electric field and also the in-plane one due to the intrinsic SOC from the contribution of the valence band due to the space inversion symmetry. However, according to the latest report by Kormányos et al.\textsuperscript{39} this intrinsic SOC from the contribution of the valence band is negligible since the splitting of the valence band is much smaller than the typical band gap. In contrast, the intrinsic SOC from the conduction band, which manifests itself as a Zeeman-like term with opposite effective magnetic fields in the two valleys, is absent in their calculation.\textsuperscript{37} The Zeeman-like term together with the intervalley electron-phonon scattering can open an intervalley spin relaxation channel, which has been shown of significant importance to the spin relaxation in both rippled single-layer graphene\textsuperscript{40} and also bilayer graphene\textsuperscript{41} Moreover, only the disorder is included in a phenomenological manner in their calculation. The electron-electron Coulomb and electron-phonon interaction, which have been demonstrated to play an important role in spin relaxation in semiconductors\textsuperscript{41} and also graphene at high temperature\textsuperscript{45} are absent.

In the present work, with the electron-electron Coulomb, (both the intra- and inter-valley) electron-phonon and the electron-impurity scatterings explicitly included, we study the electron spin relaxation due to the D’yakonov-Perel’\textsuperscript{28} (DP) mechanism in monolayer MoS\textsubscript{2}...
in the absence of external electric field by the kinetic spin Block equation (KSBE) approach. According to the latest report by Kormányos et al., the effective magnetic field of the intrinsic SOC of the conduction band is given by

$$\Omega^\mu = (0, 0, 2 \lambda \mu)$$

with the $z$-axis being perpendicular to the monolayer MoS$_2$ plane. Here, $\lambda$ and $\mu = 1(-1)$ represent the strength of the SOC and K(K') valleys, respectively. It is noted that the contribution of the hybridization by the intrinsic SOC of the valence band to the energy bands is neglected since the energy scale of this spin splitting is much smaller than the typical band separation. This effective magnetic field is oriented perpendicular to the monolayer MoS$_2$ plane, which leads to the absence of the DP spin relaxation for spins along this direction. This is similar to the case of (110) III-V zinc-blende symmetric quantum wells. In addition, the effective magnetic field is momentum independent, indicating an absence of the intravalley inhomogeneous broadening for in-plane spins. Therefore, the intravalley spin relaxation process contributed by the electron-electron Coulomb, intravalley electron-phonon and electron-impurity scatterings are irrelevant to the in-plane spin relaxation. Here, the SOC only serves as a Zeeman-like term with opposite effective magnetic fields perpendicular to monolayer MoS$_2$ plane in the two valleys. The Zeeman-like term together with the intervalley electron-phonon scattering opens an intervalley in-plane spin relaxation channel, which is similar to the case of bilayer graphene and also ripped single-layer graphene. This spin relaxation channel suppresses the in-plane SRT effectively at high temperature. We also find that this system is always in the weak intervalley scattering limit, which determines that the in-plane spin relaxation time is proportional to the intervalley momentum scattering time. As a result, a monotonic decrease of the in-plane SRT with increasing temperature is shown. Moreover, a peak is predicted in the electron-density dependence of the in-plane SRT, which results from the crossover from the nondegenerate to degenerate limit. We also show a rapid decrease of the in-plane SRT with increasing initial spin polarization at low temperature, which is very different from the previous studies in both semiconductors and single-layer graphene but similar to the case of bilayer graphene.

This paper is organized as follows. In Sec. II, we introduce our model and construct the KSBEs. Then in Sec. III, we calculate the dependences of the electron spin relaxation on temperature, electron density and initial spin polarization in monolayer MoS$_2$. We give a summary in Sec. IV.

II. MODEL AND KSBEs

With the effective mass approximation, the Hamiltonian of the conduction band near the K(K') points in monolayer MoS$_2$ can be described by

$$H^{\mu}_{\text{eff}} = \epsilon_{\mu k} + \Omega^{\mu} \cdot \sigma / 2,$$

according to the latest report by Kormányos et al. Here, $\epsilon_{\mu k} = h^2 k^2 / 2m^*$ with $k$ and $m^*$ being the in-plane momentum relative to the K(K') points and the effective mass, respectively. $\sigma$ are the Pauli matrices and $\Omega^{\mu}$ is given in Eq. (1).

We then construct the microscopic KSBEs to investigate the intrinsic electron spin relaxation in monolayer MoS$_2$. The KSBEs are given by

$$\partial_t \hat{\rho}_{\mu k} = \partial_t \hat{\rho}_{\mu k}|_{\text{coh}} + \partial_t \hat{\rho}_{\mu k}|_{\text{scat}},$$

in which $\hat{\rho}_{\mu k}$ stand for the density matrices of electrons with the diagonal terms $\rho_{\mu k, \sigma \sigma} \equiv f_{\mu k, \sigma}$ ($\sigma = \pm \frac{1}{2}$) representing the distribution functions and the off-diagonal ones $\rho_{\mu k, \sigma}(-\frac{1}{2}) = \rho_{\mu k}(-\frac{1}{2} | \frac{1}{2})$ describing the spin coherence. The coherent terms read

$$\partial_t \hat{\rho}_{\mu k}|_{\text{coh}} = -i [\Omega^{\mu} (k) \cdot \sigma / 2 + \Sigma^{\text{HF}}_{\mu k}, \hat{\rho}_{\mu k}],$$

where $[A, B] \equiv AB - BA$ is the commutator; $\Sigma^{\text{HF}}_{\mu k} = - \sum_{\mu' k'} V_{\mu k, \mu' k'}$ denote the Coulomb Hartree-Fock (HF) term, with $V_{\mu k, \mu' k'}$ representing the screened Coulomb potential. The scattering terms $\hat{\rho}_{\mu k}|_{\text{scat}}$ are the scattering terms including the electron-electron Coulomb ($\langle \mathbf{V}_{\mu k, \mu' k'}^{\mu} \rangle$), electron-impurity ($\langle \mathbf{U}_{\mu k, \mu' k'}^{\mu} \rangle$), intravalley electron-convective (AC) phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{\mu} \rangle$), electron-optical (OP) phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{\mu} \rangle$), and especially the intervalley electron-phonon scattering including the electron-KTA phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{KTA} \rangle$), electron-KLA phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{KLA} \rangle$), electron-KTO phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{KTO} \rangle$) and electron-KLO phonon ($\langle \mathbf{M}_{\mu k, \mu' k'}^{KLO} \rangle$) scatterings. Here, KTA, KLA, KTO and KLO correspond to the transverse acoustic (TA), longitudinal acoustic (LA), transverse optical (TO) and longitudinal optical (LO) phonon modes at K point, respectively. Specifically, the electron-electron and electron-impurity scattering terms can be written as

$$\partial_t \hat{\rho}_{\mu k}|_{\text{ei}} = -\pi N_e \sum_{k'} [U_{\mu k, \mu k'}^\alpha]^2 \delta (\epsilon_{\mu k'} - \epsilon_{\mu k}) \times (\hat{\rho}_{\mu k'}^\alpha \hat{\rho}_{\mu k}^\alpha - \hat{\rho}_{\mu k'}^\alpha \hat{\rho}_{\mu k}^\alpha) + \text{H.c.},$$

$$\partial_t \hat{\rho}_{\mu k}|_{\text{ee}} = -\pi \sum_{\mu' k' k''} |V_{\mu k, \mu' k'}^{\mu}|^2 \delta (\epsilon_{\mu k'} - \epsilon_{\mu k} + \epsilon_{\mu' k''})$$

$$- \epsilon_{\mu' k''} - k + k' \left[ \text{Tr} (\hat{\rho}_{\mu' k'' k'}^\alpha - \hat{\rho}_{\mu' k'' k'}^\alpha) \hat{\rho}_{\mu k}^\alpha \hat{\rho}_{\mu k}^\alpha + \text{H.c.} \right].$$

In these equations, $N_e$ stands for the impurity density; $\hat{\rho}_{\mu k}^\alpha \equiv 1 - \hat{\rho}_{\mu k}$, $\hat{\rho}_{\mu k}^\alpha \equiv \hat{\rho}_{\mu k}$. As for the electron-phonon scattering terms, their detailed expressions can be found in Ref. The scattering matrix elements

$$|V_{\mu k, q - \mathbf{Q}}^{\mu}|^2 = \left( \frac{q}{Q} \right)^2,$$
and

\[ |U_{k,k-q}^{\mu}|^2 = Z_i^2 |V_{k,k-q}^{\mu}|^2 \]

with \( Z_i \) being the impurity charge number (assumed to be 1 in our calculation). The quantity

\[ \varepsilon(q) = 1 - V_q^{(0)} \sum_{\mu\nu k} f_{\mu k}^{\nu} f_{k+q \mu k}^{\nu} \]

stands for the screening under the random phase approximation (RPA). \( V_q^{(0)} = 2e^2/(\pi q) \) is the two-dimensional bare Coulomb potential with \( \kappa \) being the relative static dielectric constant. The electron-phonon scattering matrix elements \( |M_{\mu k,\mu' k'}|^2 \) (\( \lambda = \text{AC, OP, KTA, KLO} \)) are explicitly given by the latest reports by Kaasbjerg et al.\footnote{22,23} Specifically, we lay out the matrix elements of the electron-KTA and -KLO phonon scatterings, which play a dominant role in the in-plane spin relaxation as will be shown later.

\[
|M_{\mu k,\mu' k}^{\text{KTA}}|^2 = \frac{\hbar^2 (D_{k,T A}^{1})^2 q^2}{2A \rho \Omega_{k,TA}} \delta_{\mu' - \mu},
\]

\[
|M_{\mu k,\mu' k}^{\text{KLO}}|^2 = \frac{\hbar^2 (D_{k,LO}^{0})^2 v^2}{2A \rho \Omega_{k,LO}} \delta_{\mu' - \mu},
\]

where \( A \) is the area of the sample; \( \rho \) is the mass density of the monolayer MoS\(_2\); \( \Omega_{k,TA} \) and \( \Omega_{k,LO} \) are the energies of KTA and KLO phonon modes, respectively; \( D_{k,TA}^{1} \) (\( D_{k,LO}^{0} \)) is the first- (zeroth-) order deformation potential corresponding to the electron-KTA (-KLO) phonon scattering; \( q = |k - k'| \). It is noted that the screening by the carriers is included in the electron-phonon scattering in their works with the Thomas-Fermi (Debye-Hückel) screening in the degenerate (nodegenerate) limit.\footnote{22,23} This screening is also taken into account in the electron-phonon scattering in the present work but with the RPA one, which reproduces the Thomas-Fermi and Debye-Hückel ones in the degenerate and nodegenerate limits, respectively.\footnote{27,28}

### III. NUMERICAL RESULTS

As reported, there still remain controversies over the band structure of monolayer MoS\(_2\),\footnote{22,23,27,32,34,38,39} where different energy gaps, effective masses and the spin splittings are given. Here, we take the effective mass and the strength of the SOC being \( m^* = 0.48m_0 \)\footnote{22,23} and \( \lambda = 1.5 \) meV,\footnote{37,39} respectively. \( m_0 \) stands for the free electron mass. The relative static dielectric constant is chosen to be \( \kappa = 3.43 \).\footnote{22} The mass density \( \rho = 3.1 \times 10^{-7} \) g/cm\(^2\); the KTA and KLO phonon energies are \( \Omega_{k,TA} = 23 \) meV and \( \Omega_{k,LO} = 42 \) meV, respectively; the deformation potentials for the KTA and KLO phonons are \( D_{k,TA} = 5.9 \) eV and \( D_{k,LO} = 2.6 \times 10^{8} \) eV/cm, respectively.\footnote{27,37} With these parameters, our results are obtained by numerically solving the KSBEs [Eq. (3)\footnote{44}]. Specifically, we calculate the time evolution of spin polarization along \( \mathbf{n} \), i.e.,

\[ P(t) = \sum_{\mu k} \text{Tr}[\hat{\rho}_{\mu k}(t) \sigma \cdot \mathbf{n}] / N_e \]

with \( N_e \) representing the electron density. Then the SRT can be obtained from the slope of the envelope of the spin polarization.\footnote{44} Note that the initial spin polarization is set to be 2.5\% and the spin-polarization direction is chosen in the monolayer MoS\(_2\) plane unless otherwise specified.

#### FIG. 1: (Color online) Total in-plane SRT \( \tau_s \) (\( \times \)) and that calculated with only the electron-KTA phonon (\( \blacksquare \)) or electron-KLO phonon (\( \bullet \)) scattering included as function of temperature \( T \). In the calculation, \( N_e = 7 \times 10^{12} \) cm\(^{-2}\).

##### A. Temperature dependence of spin relaxation

We first study the temperature dependence of the spin relaxation. The in-plane SRT \( \tau_s \) as function of temperature \( T \) is plotted in Fig. 1. It is seen that the SRT decreases monotonically with the increase of the temperature. To understand this behavior, we calculate the temperature dependence of the SRT with only the electron-electron Coulomb, electron-impurity, intravalley electron-AC-phonon, electron-OP-phonon, or intervalley electron-phonon scatterings, which only contribute to the intravalley spin relaxation channel, are negligible due to the absence of a momentum-dependent effective magnetic field of the SOC [Eq. (1)\footnote{44}]. Therefore, the SRT is only determined by the intervalley spin relaxation channel induced by the intervalley electron-phonon scattering together with the Zeeman-like term with opposite effective magnetic fields in the two valleys. This is similar to the case of rippled single-layer graphene\footnote{22} and also bi-
layer graphene. With the Zeeman-like term \((0, 0, 2\lambda \mu)\) [Eq. \(1\)], the in-plane SRT is given by
\[
\tau_s = \left\{ \begin{array}{ll}
\tau_v & \text{weak scattering (}2\lambda \tau_v \geq 1) \\
\frac{1}{2\lambda \tau_v} & \text{strong scattering (}2\lambda \tau_v \ll 1) \end{array} \right.
\]
(12)
according to the report by Zhang et al.\(^{42}\) Here, \(\tau_v\) represents the intervalley electron-phonon scattering time. It is noted that the intervalley electron-phonon scattering is always in the weak scattering limit for the electron density up to \(2 \times 10^{13}\) cm\(^{-2}\), therefore \(\tau_s = \tau_v\) throughout our work. As a result, the SRT decreases with the enhancement of the intervalley electron-phonon scattering as the temperature increases.

In addition, we also show a comparison of contributions from two leading intervalley electron-phonon scatterings, i.e., electron-KTA and electron-KLO ones in Fig. 1. We find that the electron-KTA phonon scattering plays a more important role in spin relaxation at low temperature whereas the electron-KLO phonon scattering becomes more important at high temperature. This can be understood from the contribution of the electron-phonon scattering matrix element together with phonon number. It is noted that the electron-KTA phonon scattering matrix element is smaller than the electron-KLO phonon one whereas the KTA phonon mode has a larger phonon number due to a smaller phonon energy.\(^{22,34}\) At low temperature, the phonon number of KTA mode is much larger than that of KLO mode, which makes the electron-KTA phonon scattering stronger and hence more important to the spin relaxation. However, at high temperature, the phonon numbers of two modes become comparable. Then the electron-KLO phonon scattering plays a more important role in spin relaxation due to the larger scattering matrix element. It is noted that the contributions of the electron-KLA and -KTO phonon scatterings to the spin relaxation are marginal.

B. Electron-density dependence of spin relaxation

Then we turn to investigate the electron-density dependence of the in-plane SRT and show the results at \(T = 200\) K in Fig. 2. We find that the SRT first increases then decreases with increasing electron density. By comparing the SRT calculated with and without the screening shown in Fig. 2, one determines that the peak originates from the screening. Specifically, at low electron density, the system is in the nondegenerate limit where the screening is given by
\[
\varepsilon(q) = 1 + N_e V_q^{(0)}/(\pi \hbar^2)
\]
(13)
approximately with \(k_B\) being the Boltzmann constant. Also in this limit, \(V_q^{(0)} \propto q^{-1}\) is insensitive to the electron density. Therefore, the screening increases as the electron density increases, leading to a decrease of the intervalley electron-phonon scattering. As a result, the SRT \((\tau_s = \tau_v)\) increases with increasing electron density. When the electron density further increases, the system turns to the degenerate limit with the screening being
\[
\varepsilon(q) = 1 + m^* V_q^{(0)}/(\pi \hbar^2)
\]
(14)
approximately. In this limit, \(V_q^{(0)} \propto q^{-1} \sim q_F^{-1} \propto N_e^{-1/2}\), indicating that the screening decreases with the increase of the electron density. Therefore, the intervalley electron-phonon scattering is enhanced, leading to a decrease of the SRT with increasing electron density. As a result, a peak is observed in the electron-density dependence of the SRT when the system changes from the nondegenerate to degenerate limit. In addition, we show the SRT calculated with only the electron-KTA or -KLO phonon scattering included in Fig. 2. It is seen that both show peaks in the electron density dependence. As for the electron-KLA and -KTO phonon scatterings, they are negligible to the spin relaxation similar to the case of the temperature dependence mentioned above.

![FIG. 2: (Color online) Total in-plane SRT \(\tau_s (\times)\) and that calculated with only the electron-KTA (■) or electron-KLO (●) phonon scattering included as function of the electron density \(N_e\). The dashed curve stands for the calculation without the screening. In the calculation, \(T = 200\) K.](image)

C. Initial spin polarization dependence of spin relaxation

In the previous investigations in both semiconductor systems\(^{53-56}\) and single-layer graphene,\(^{43}\) a significant increase of the SRT is shown with the increase of the initial spin polarization, which originates from the contribution of the Coulomb HF term. However, in the case of bilayer graphene, the SRT decreases rapidly with increasing initial spin polarization at low temperature whereas shows a mild increase at high temperature.\(^{43}\) In this case, the SRT is determined by the intervalley
electron-phonon scattering. Here, we also investigate the initial spin polarization dependence of spin relaxation. In Fig. 3, we plot the in-plane SRT as function of the initial spin polarization \( P \) at \( T = 100 \) (300) K. We find that the SRT shows a rapid decrease with increasing \( P \) at \( T = 100 \) K whereas decreases mildly with the increase of \( P \) at \( T = 300 \) K. We first focus on the case of \( T = 100 \) K. The rapid decrease of the SRT at \( T = 100 \) K is very different from the previous studies in both semiconductors\(^{43, 45, 53, 55, 56} \) and single-layer graphene\(^{53, 56} \) but similar to the case of bilayer graphene at low temperature.\(^{45, 53}\) The underlying physics is understood as follows. The SRT is also determined by the intervalley electron-phonon scattering whereas the contribution of the Coulomb HF term is negligible to the spin relaxation by comparing the calculation with and without the Coulomb HF term shown in Fig. 3. The interval valley electron-phonon scattering is in the weak interval valley scattering limit, i.e., the SRT \( \tau_s = \tau_v \) [Eq. (12)]. As a result, the SRT decreases significantly with the increase of the initial spin polarization due to the enhancement of the interval valley electron-phonon scattering.\(^{45}\) As for the case of \( T = 300 \) K, the decrease of the SRT is different from the cases of semiconductors\(^{43, 45, 53, 56} \) and bilayer graphene.\(^{53, 56}\) Here, the SRT is also determined by the interval valley electron-phonon scattering and this scattering is in the weak scattering limit, i.e., the SRT \( \tau_s = \tau_v \) [Eq. (12)]. This makes the SRT show the same trend as the case of \( T = 100 \) K. A mild decrease of the SRT originates from the insensitivity of \( \tau_v \) to the initial spin polarization at \( T = 300 \) K.\(^{43}\)

IV. SUMMARY

In summary, we have investigated the intrinsic electron spin relaxation due to the DP mechanism in monolayer MoS\(_2\). The effective magnetic field of the intrinsic SOC is perpendicular to monolayer MoS\(_2\) plane, which leads to the absence of the DP spin relaxation for spins along this direction. In addition, this effective magnetic field is momentum independent, indicating that the interval valley spin relaxation channel is absent for the in-plane spins. However, this SOC supplies a Zeeman-like term with opposite effective magnetic fields perpendicular to the monolayer MoS\(_2\) plane in the two valleys, which is similar to the case in rippled single-layer graphene and also bilayer graphene. This Zeeman-like term, together with the interval valley electron-phonon scattering, opens an interval valley relaxation channel for in-plane spin polarization, which has not been yet reported in the previous literature on monolayer MoS\(_2\). This spin relaxation channel can markedly suppress the in-plane SRT at high temperature. In addition to the interval valley electron-phonon scattering, the electron-impurity, electron-electron Coulomb and intravalley electron-phonon scatterings are also included to calculate the in-plane SRT. However, the contribution of these scatterings to in-plane spin relaxation is negligible due to the absence of intravalley relaxation process. Moreover, we find that the interval valley scattering is always in the weak scattering limit in this material. Therefore, the SRT is always proportional to the interval valley momentum scattering time. This results in a monotonic decrease of the in-plane SRT with the increase of temperature. In addition, a peak is predicted in the electron density dependence of the in-plane SRT, which originates from the crossover from the nondegenerate to degenerate limit. We also find that the in-plane SRT decreases rapidly with the increase of the initial spin polarization at low temperature. This is very different from the previous studies in both semiconductors and single-layer graphene whereas similar to the case of bilayer graphene.

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