Spin and Valley Noise in Two-Dimensional Dirac Materials

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We develop a theory for optical Faraday rotation noise in two-dimensional Dirac materials. In contrast to spin noise in conventional semiconductors, we find that the Faraday rotation fluctuations are influenced not only by spins but also the valley degrees of freedom attributed to intervalley scattering processes. We illustrate our theory with two-dimensional transition metal dichalcogenides and discuss signatures of spin and valley noise in the Faraday noise power spectrum. We propose optical Faraday noise spectroscopy as a technique for probing both spin and valley relaxation dynamics in two-dimensional Dirac materials.

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Introduction.– The discovery of two-dimensional (2D) Dirac materials has been a rapidly unfolding trend recently [1]. Materials such as h-BN [2] and transition metal dichalcogenides, in particular MoS$_2$ semiconductor [3], hold great promise for electronic and optical applications [4]. An important feature of these Dirac semiconductors is their intrinsic valley degrees of freedom that can couple to magnetic perturbations. 2D Dirac materials are also promising candidates for implementing spintronic devices due to their potential in achieving long electron spin life-time and achieve non-dissipative control of spin and valley currents via the quantum spin/valley Hall effect [5]. However, the spin dynamics of near-equilibrium Dirac electrons in 2D MoS$_2$ is difficult to explore with a standard optical pump-probe setup because of strong excitonic effects [6].

The recently developed optical spin noise spectroscopy (SNS) [7] is an alternative technique to explore spin dynamics in semiconductors. It has been used successfully to determine the spin coherence and spin relaxation times in bulk GaAs [8], quantum wells [9,10], and quantum dots [11]. Spin noise is studied via illumination of a linearly polarized light on a mesoscopic region of the sample. The Faraday rotation angle of the polarization axis of the measurement beam is proportional to the instantaneous total spin polarization and can be detected with a sensitivity reaching a single spin level [12] and picosecond time resolution [13]. The noise power spectrum typically exhibits a peaked profile in a magnetic field transverse to the measurement axis, with the peak location yielding the g-factor and the broadening yielding the spin relaxation time. Importantly, SNS is a minimally invasive approach that does not require to artificially create a strongly nonequilibrium spin polarization.

In this Letter, we develop a theory of the optical spin noise spectroscopy for two-dimensional Dirac semiconductors. We take 2D MoS$_2$ as the prototypical example and study its spin noise dynamics within a Langevin equation framework. We identify and discuss regimes of interest where fluctuations due to spin flip and intervalley scattering events predict measurable signatures in the Faraday noise power spectrum from which information about spin and valley relaxation processes can be inferred. Our main finding is that the Faraday rotation noise of Dirac electrons is sensitive to fluctuations in both spin and valley degrees of freedom. Since the noise contribution from spins is sensitive to an external magnetic field, it should be possible to clearly separate spin and valley dynamics.

Faraday Rotation Fluctuations.– We first establish a general relationship between the Faraday rotation fluctuations and the spin noise in a 2D system of degenerate itinerant electrons. To this end, we define the positive and negative helicity components of the optical conductivity tensor as $\sigma_{\pm} = \sigma_{xx} \pm i\sigma_{xy}$, where $\sigma_{xx}$ and $\sigma_{xy}$ are the longitudinal and Hall conductivities of the 2D system. The real part $\text{Re}\sigma_{\pm} = \text{Re}\sigma_{xx} \mp \text{Im}\sigma_{xy}$ corresponds to dissipative on-shell electronic transitions whereas the imaginary part $\text{Im}\sigma_{\pm} = \text{Im}\sigma_{xx} \pm \text{Re}\sigma_{xy}$ corresponds to dissipationless virtual transitions. Throughout this Letter we adopt ‘natural units’ by normalizing optical conductivities by the speed of light $c$ so that $\sigma_{xx},\sigma_{xy}$ are in units of the fine structure constant $\alpha \approx 1/137$. Analysis of the electromagnetic transmission problem through the 2D layer [14] yields the Faraday rotation $\theta_F = (\theta_+ - \theta_-)/2$, where $\theta_{\pm} = -\tan^{-1}[2\pi\text{Im}\sigma_{\mp}/(1 + 2\pi\text{Re}\sigma_{\mp})]$ is the phase of the positive and negative helicity component, respectively, of the electric field transmitted through the layer. It is clear that $\text{Im}\sigma_{\pm}$ contributes to circular birefringence and $\text{Re}\sigma_{\pm}$, in subleading orders, to circular dichroism.

The power of the optical spin noise spectroscopy lies in its non-perturbative nature that allows it to probe the system under study at thermal equilibrium. This is achieved by tuning the light frequency to a value that is smaller than the electronic band gap $E_g$, with the detuning $\omega_d \ll E_g$ so as to maximize the observed Faraday rotation signal. Dissipative transitions are therefore forbidden with $\text{Re}\sigma_{\pm} \approx 0$, and the optical response is largely reactive and dissipationless.

2D materials obeying the massive Dirac energy dispersion are described by the Hamiltonian $H_0 = \hbar v (\tau k_x \hat{\sigma}_x + k_y \hat{\sigma}_y) + (\Delta/2)\hat{\sigma}_z$, where $v$ is the band velocity, $\Delta$ the band gap, $\hat{\sigma}_x$ are the Pauli matrices describing the sublattice degrees of freedom, and $\tau = \pm 1$ denotes
the valley degrees of freedom $K, K'$. The valley degrees of freedom in addition to spins are endowed with magnetic moments that can couple to time-reversal breaking perturbations such as external magnetic fields or circularly polarized light. Therefore the Faraday rotation will be sensitive to the electron population differences between spins and valleys.

To derive the Faraday rotation, we consider the situation with a nonequilibrium distribution of Fermi levels among the four possible quantum states $(K, \uparrow), (K', \uparrow), (K', \downarrow), (K, \downarrow)$ in the conduction bands of the system, where $\uparrow, \downarrow$ denote the up and down spins. Intra-valley spin-conserving scattering is fast compared to intra-valley spin-flip or inter-valley scattering, so one can write the corresponding Fermi levels for each spin and valley as $\epsilon_{F,s} = \epsilon_F + \delta\epsilon_{F,s}^\tau$, where $\tau = \pm 1$ corresponds to $K, K'$ and $s = \pm 1$ to $\uparrow, \downarrow$. $\epsilon_F$ is the equilibrium Fermi energy measured from the conduction band edge, and $\delta\epsilon_{F,s}^\tau$ is the nonequilibrium Fermi energy fluctuations for each spin $s$ and valley $\tau$. Such a nonequilibrium distribution of Fermi energies is constrained by conservation of the total number of electrons, therefore $\delta\epsilon_{F,s}^\tau$ must satisfy an implicit condition given by $\sum_{s, \tau = \pm 1} \delta n_{F,s}^{\tau} = 0$ where $\delta n_{F,s}^{\tau}$ is the density fluctuations corresponding to $\delta\epsilon_{F,s}^\tau$.

The optical conductivity due to such a nonequilibrium Fermi level distribution is

$$\delta\sigma_{\pm} = \sum_{\tau = K, K'} \sum_{s = \pm 1} \sigma_{\tau, s} \left( \epsilon_F + \delta\epsilon_{F,s}^\tau \right),$$

where $\sigma_{\tau, s}$ refers to the conductivity for spin $s$ and valley $\tau$ with the frequency label $\omega$ suppressed for clarity. We expand Eq. (1) up to first order in the small parameter $\delta\epsilon_{F,s}^\tau/\epsilon_F$ as appropriate for small fluctuations. Noting that the two Hall contributions at the Fermi level $\epsilon_F$ due to the $K, K'$ valleys cancel and using the expressions for $\theta_{\pm}$, we obtain the nonequilibrium Faraday rotation

$$\theta_F = \frac{2\pi}{1 + \frac{2\pi}{2} \text{Im} \sigma_{xx}(\epsilon_F)|_{\epsilon_F = \epsilon_F}^2} \sum_{\tau = K, K'} \sum_{s = \pm 1} \frac{\partial \text{Re} \sigma_{\tau, s}(\epsilon)}{\partial \epsilon} \delta\epsilon_{F,s}^\tau,$$

where $\sigma_{xx}$ is the total longitudinal conductivity summed over all spins and valleys.

2D transition metal dichalcogenides. Eq. (2) is applicable to massive Dirac fermion 2D systems or their multilayered derivatives. In particular, the large energy gap $E_g \sim eV$ of 2D transition metal dichalcogenides makes them especially suitable as prototypical 2D Dirac materials amenable to study with SNS. Because of spin-orbit coupling, 2D transition metal dichalcogenides also exhibit coupled spin and valley dynamics. The low-energy $K, \delta$ Hamiltonian near the Brillouin zone corners $K, K'$ is given by $H = H_0 + H_{SO}$, where $H_0$ is the massive Dirac Hamiltonian defined earlier and $H_{SO} = -\lambda \tau (\hat{s}_z - 1) \hat{s}_z/2$ is the spin-orbit coupling term with $\hat{s}_z$ the Pauli matrix describing the spin degrees of freedom and strength $\lambda \sim 10 - 100$ meV.

With the band gap given by $E_g = \Delta - \lambda$, the frequency of the probe laser beam is $\omega = \Delta - \lambda - \omega_d$. The optical Hall and longitudinal conductivities are calculated using the Kubo formalism and we find

$$\text{Re} \sigma_{\tau, s} = \frac{-\tau \alpha}{4\pi} \left( \Delta - s \tau \lambda \right) F_{\tau}^\tau(\epsilon_F, \omega_d),$$

$$\text{Im} \sigma_{xx,s} = -\frac{\alpha}{8\pi} \left\{ \frac{2\epsilon_F + \Delta - s \tau \lambda}{\Delta - \lambda - \omega_d} \left[ 1 + \frac{\Delta - s \tau \lambda}{2\epsilon_F + \Delta - s \tau \lambda} \right]^2 \right\} \left[ 1 + \frac{\Delta - s \tau \lambda}{\Delta - \lambda - \omega_d} \right]^2 F_{\tau}^\tau(\epsilon_F, \omega_d),$$

for each spin and valley $s, \tau = \pm 1$ and $F_{\tau}^\tau(\epsilon_F, \omega_d) = \ln[2\epsilon_F - (s - 1)\lambda + \omega_d]/[2\epsilon_F + 2\Delta - (s + 1)\lambda - \omega_d]$. Equations (3)-(4) are valid for our regime of interest at low temperatures $k_B T \ll \epsilon_F$ and for clean enough samples such that disorder broadening $\hbar/\tau \ll \omega_d$.

To connect the Faraday rotation fluctuations with experimental observables, we first express Eq. (2) in terms of electron densities. The Fermi level fluctuations can be related to the number density fluctuations as $\delta n_{F,s}^\tau = \nu_{F,s}^\tau(\epsilon_F) \delta\epsilon_{F,s}^\tau$, where $\nu_{F,s}^\tau(\epsilon) = (2\epsilon + \Delta + s \tau \lambda)/4\pi(hv)^2$ is the density of conduction band states per unit area. The nonequilibrium spin density fluctuations are given by $\delta n_{\tau}^s = (\delta n_{F,s}^{\uparrow} - \delta n_{F,s}^{\downarrow})/2$ for valley $K$ and similarly for $K'$, whereas the valley density fluctuations are given by $\delta n_K = (\delta n_{\tau}^{\uparrow} + \delta n_{\tau}^{\downarrow})/2$, with $\delta n_{K'} = -\delta n_K$. Using these relations and substituting Eqs. (3)-(4) in Eq. (2), we obtain the Faraday rotation fluctuations

$$\theta_F = \mathcal{L}_\tau S_\tau + \mathcal{L}_v N_v,$$

where $S_\tau = (\delta n_{\tau}^K + \delta n_{\tau}^{K'})A$ and $N_v = (\delta n_K - \delta n_{K'})A$ are respectively the total spin and total valley polarization fluctuations over the cross-sectional area $A$ of the incident probe laser beam, and $\mathcal{L}_\tau = (\mathcal{L}_\tau \pm \mathcal{L}_v)/A$ are the spin and valley coupling coefficients respectively with $\mathcal{L}_\tau = \pm 8\pi\alpha(hv)^2(\Delta \mp \lambda)/(\omega_d^2 - (2\epsilon_F + \Delta \mp \lambda)^2)(2\epsilon_F + \Delta \mp \lambda)[1 + 2\pi\text{Im} \sigma_{xx}(\epsilon_F)|_{\epsilon_F = \epsilon_F}^2]$. Equation (5) is a central result of this Letter. Importantly, valley fluctuations and spin fluctuations contribute on an equal footing to the Faraday rotation in Dirac materials. This implies that the relaxation dynamics of both spins and valleys can be optically probed using Faraday rotation spectroscopy.

To determine the noise properties of the Faraday rotation, we now derive the kinetic equations governing the spin and valley fluctuations. To this end, we first obtain an effective Hamiltonian for conduction band states near the band edge. Using Löwdin’s partitioning [16] we find the following effective Hamiltonian for conduction band electrons with Fermi level $\epsilon_F \ll \Delta$ in an external field:

$$H_c = \frac{\hbar^2 k^2}{2m_e} + \frac{\hbar \Omega_{SO}(k)}{2} \hat{s}_z + \tilde{g}(k) \mu_B \mathbf{B} \cdot \hat{s},$$

where $\Omega_{SO}(k)$ is the spin-orbit coupling constant, and $\tilde{g}(k)$ is the effective g-factor.
where \( m_e = (\Delta^2 - \lambda^2)/(2\Delta u^2) \) is the effective mass, \( g(k) = g[1 + (vk)^2]/(\Delta^2 - \lambda^2) \) is the renormalized g-factor and \( \Omega_{SO}(k) = 2\lambda h(k)^2/(\Delta^2 - \lambda^2) \). Note that the spin-orbit coupling acts on conduction electrons as an effective out-of-plane magnetic field with opposite sign \( \tau = \pm \) in different valleys.

Using Eq. (6) and the equation of motion for the spin-valley density matrix [17], we obtain the following kinetic equations for the total spin fluctuations per valley \( S^\tau \) and total valley polarization fluctuations \( N_v \):

\[
\frac{\partial S^\tau}{\partial t} + \tau S^\tau \times \Omega_{SO} + S^\tau \times \Omega_L = -\frac{S^\tau}{T_S} + \frac{S^{\tau - \delta S^\tau}}{T_v} + \eta_s S^\tau, \tag{7}
\]

\[
\frac{\partial N_v}{\partial t} = -\frac{N_v}{T_v} + \eta^N, \tag{8}
\]

where \( \Omega_{SO} \) is the effective out-of-plane magnetic field induced by spin-orbit coupling and \( \Omega_L = \hat{g}(k_f)\mu_B B/h \) at the Fermi level. The spin relaxation time \( T_S [18] \) captures the relaxation of nonequilibrium spin fluctuations in one valley due to possible spin-flip scattering with magnetic defects [19] as well as D’yakonov-Perel’ and Elliot-Yafet mechanisms [20]; whereas the valley relaxation time \( T_v \) describes the relaxation of valley fluctuations due to inter-valley scattering from atomically-sized non-magnetic impurities or electron-phonon interaction that induce momentum transfer on the order of inverse lattice spacing [21]. \( \eta_s^\tau \) and \( \eta_s^\tau \) are the corresponding noise sources that describe spin fluctuations \( S \) in valley \( \tau \) due to spin and valley relaxation respectively, while \( \eta^N \) describes fluctuations of the valley polarization \( N_v \). The different physical origin of these noise terms implies that \( \eta_s^\tau \) is uncorrelated to \( \eta_s^\tau \) and \( \eta^N \). The latter can also be considered mutually uncorrelated if inter-valley scattering processes of up and down spins are statistically independent and equally probable. Due to the large (mesoscopic) number of electrons involved, the time correlators of these noise sources at thermodynamic equilibrium can be regarded as \( \delta \)-function correlated, and are constrained by the Fluctuation-Dissipation Theorem to have the following form [17, 23]:

\[
\langle (\eta_s^{\tau}(t))_\alpha(\eta_s^{\tau}(t'))_\beta \rangle = \delta_{\tau\tau'} \delta_{\alpha\beta} \frac{2DkB_T}{T_S} \delta(t-t'), \tag{9}
\]

\[
\langle (\eta_s^{\tau}(t))_\alpha(\eta_s^{\tau}(t'))_\beta \rangle = \frac{2DkB_T}{T_v} \delta(t-t'), \tag{10}
\]

\[
\langle \eta^N(t)\eta^N(t') \rangle = \frac{4DkB_T}{T_v} \delta(t-t'), \tag{11}
\]

where \( \alpha, \beta = x, y, z, T \) is temperature and \( D \) is the density of conduction band states at the Fermi surface in the observation area \( A \) per band and per unit energy.

The Faraday rotation noise power follows from Eq. (5):

\[
\langle |\delta_{F}(\omega)|^2 \rangle = L_s^2 \langle |\tilde{S}_z(\omega)|^2 \rangle + L_v^2 \langle |\tilde{N}_v(\omega)|^2 \rangle, \tag{12}
\]

where \( \tilde{S}_z(\omega) \) and \( \tilde{N}_v(\omega) \) are the total spin and total valley polarization noise power with \( X(\omega) \equiv \lim_{T \to \infty} \int_0^T dt e^{i\omega t} X(t) \) where \( T_m \) is the measurement time. Since \( \eta^N, \eta_s^\tau, \eta_s^\tau \) are mutually uncorrelated, the dynamics of the valley polarization \( N_v \) decouples from that of the spins and can be readily obtained as

\[
\langle |\tilde{N}_v(\omega)|^2 \rangle = \frac{4DkB_T/T_v}{\omega^2 + 1/T_v^2}. \tag{13}
\]

The valley dynamics is therefore described by a single Lorentzian noise power peak centered at zero frequency that is insensitive to the applied magnetic field, and the valley relaxation time \( T_v \) can be extracted from the width of the peak. For the spin dynamics, the Langevin equation for spins Eq. (7) corresponds to an Ornstein-Uhlenbeck process [23] with the spin noise correlators given by

\[
\langle \tilde{S}_s^\alpha(\omega)\tilde{S}_s^{\tau'}(-\omega) \rangle = \left( \frac{1}{A} - i\omega \frac{G}{A + i\omega} \right)_{\tau\tau'}, \tag{14}
\]

where \( A \) is the relaxation time matrix \( A_{\tau,\tau'} = -\delta_{\alpha\beta}(\delta_{\tau\tau'}/T_S + (\delta_{\tau\tau'} - \delta_{\tau',\tau})/T_v) + \delta_{\tau\tau'} (\tau\Omega_{SO} \epsilon_{\alpha\beta} + \Omega_{L} \epsilon_{\alpha\beta}) \) (here \( \epsilon_{ijk} \) is the Levi-Civita symbol) and \( G_{\tau,\tau'} \) is defined by Eq. (14). Equation (14) can be evaluated analytically in closed form [24].

The 2D transition metal dichalcogenide MoS2 has a band gap in the optical spectrum \( \Delta = 1.7 \) eV and spin-orbit coupling \( \lambda = 75 \) meV [15]. For typical Fermi energies \( \epsilon_F \sim 10 \) meV near the band edge, detuning \( \omega_d \sim 10 \) meV, and laser spot size \( A \sim 1 \) mm$^2$, we have total spin and valley polarization fluctuations \( \sqrt{\langle |S_z|^2 \rangle}, \sqrt{\langle |N_v|^2 \rangle} \sim 200 \) in the observation region with a total number of electrons \( \sim 10^8 \). The Faraday rotation fluctuations is approximately equally coupled to the
valley and spin degrees of freedom with |\mathcal{L}_- - \mathcal{L}_+| \approx 0.1 and the root-mean-squared Faraday angle fluctuations \sqrt{\langle \theta_F^2 \rangle} \sim 1 \mu\text{rad}, which is within the state-of-the-art measurement capabilities.

In MoS\textsubscript{2}, the spin-orbit splitting \hbar\Omega_{SO} \approx \epsilon_F \lambda / \Delta \approx 0.5 \text{meV} corresponds to a strong magnetic field of \approx 8 \text{T}, which strongly favors out-of-plane spin alignments and hence resists Larmor precession due to the applied in-plane field. Given that the spin relaxation time is expected to be long \tau_s \sim 10 \text{ns} \cite{22}, the behavior of the noise spectrum will then critically depend on the valley scattering time. Inter-valley scatterings are seen by electron spins as random sign changes of the spin-orbit field \Omega_{SO}, so depending on the scattering rate this field may or may not be effectively averaged to zero. Our studies of Eq. (14) have identified three basic regimes \cite{21}:

(i) Slow inter-valley scattering: \(1/\tau_v \ll \Omega_{SO}\). In this regime, the Faraday rotation noise power spectrum consists of the valley noise component given by Eq. (13) and spin noise components (Fig. 1). At small fields \Omega_L \ll \Omega_{SO}, the spin noise part of the power spectrum is a Lorentzian peak centered at zero frequency

\[
\langle |\tilde{S}_z(\omega)|^2 \rangle = \frac{4Dk_B T/\tau_v}{\omega^2 + 1/\tau_v^2}.
\]

A finite frequency peak starts to emerge at \Omega_L \sim \Omega_{SO} [Fig. 1(a)]. It is centered near the effective spin precession rate \Omega_{eff} = \sqrt{\Omega_L^2 + \Omega_{SO}^2} - \Omega_{SO}. At \Omega_L > \Omega_{SO}, the total noise power clearly consists of two peaks: one centered at zero frequency due to inter-valley scattering and the finite frequency peak due to spin precession [Fig. 1(b)]. In MoS\textsubscript{2}, \Omega_{SO} \sim \text{THz} and the finite frequency peak cannot be resolved with currently accessible 100GHz bandwidth spectroscopy \cite{13}.

(ii) Moderately fast inter-valley scattering: \(1/\tau_v \lesssim \Omega_{SO}\). In this case, the finite frequency peak still appears only at large external fields, \Omega_L \sim \Omega_{SO}. However, even a moderate in-plane magnetic field (\lesssim 1 \text{T}) induces a Dyakonov-Perel-type spin relaxation by making the directions of the total field \Omega_{SO} + \Omega_L in \text{K} and \text{K}' valleys non-collinear with each other. This results in a broadening of the zero-frequency peak as shown in Fig. 2(a). The profile is generally non-Lorentzian, for which we have derived the relative amplitude of the correlator at zero and at finite magnetic fields as

\[
\langle |\tilde{S}_z(\omega = 0)|^2 \rangle_{\Omega_L = 0}/\langle |\tilde{S}_z(\omega = 0)|^2 \rangle_{\Omega_L \neq 0} = 1 + \Omega_{SO}^2 T/\Omega_{SO}^2 T_v,
\]

where \langle |\tilde{S}_z(\omega = 0)|^2 \rangle_{\Omega_L = 0}. By measuring the evolution of the peak maximum with a changing external field, one can therefore obtain the combination of parameters \Omega_L^2/(\Omega_{SO}^2 T_v) in addition to the spin relaxation time \tau_v.

(iii) Fast inter-valley scattering: \(1/\tau_v > \Omega_{SO}\). In this case, the out-of-plane spin-orbit field \Omega_{SO} is quickly randomized. A moderate magnetic field is then sufficient to drive Larmor precession which displaces the spin noise peak from zero frequency to \Omega_{L} \sim \text{GHz}, as shown in Fig. 2(b). We find that the peak is then approximately Lorentzian near the peak center \Omega_L with a spin relaxation time being renormalized by the fluctuating spin-orbit coupling \(1/\tau_v = 1/\tau_v + \Omega_{SO}^2 T_v/4\). This behavior can be observed with the state-of-the-art high bandwidth spin noise spectroscopy \cite{13}.

In conclusion, we showed that the Faraday rotation noise of Dirac electrons is sensitive to fluctuations of both the spin and valley degrees of freedom. The noise power spectrum contains an additional peak centered at zero frequency that is due to valley noise and does not couple to external in-plane magnetic fields. We also predict that, due to spin-orbit splitting of electronic bands, a Larmor peak appears only in relatively strong external magnetic fields and its width depends on the spin-orbit splitting and inter-valley scattering rate. A moderate magnetic field \sim 0.1 - 1 \text{T} is sufficient to strongly broaden the spin noise peak centered at zero frequency. If spin relaxation time is longer than 1 ns, this effect should be observable even without resorting to more complex high-bandwidth spin noise spectroscopy.

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