Strong intervalley correlation induced a magnetic order transition in monolayer MoS$_2$

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In this work, we study a model for monolayer molybdenum disulfide with including the intravalley and intervalley electron-electron interaction. We solve the model at a self-consistent mean-field level and get three solutions $L_0$, $L_1$, and $L_-$. As for $L_0$, the spin polarizations are opposite at $K$ and $K'$ valley and the total magnetization is zero. $L_\pm$ describe two degenerate spin-polarized states, and the directions of polarization are opposite for the states of $L_+$ and $L_-$. Based on these results, the ground state can be deduced to be spin polarized in domains in which their particular states can be randomly described by $L_+$ or $L_-$. Therefore, a zero net magnetization is induced for zero external magnetic field $\mathbf{B}$, but a global ferromagnetic ground state for a nonzero $\mathbf{B}$. We estimate the size of domains as several nanometers. As the increase of the chemical potential, the ground state changes between $L_0$ and $L_\pm$, indicating first order phase transitions at the borders, which is coincident with the observation of photoluminescence experiments in the absence of the external magnetic field [J. G. Roch et al., Phys. Rev. Lett. 124, 187602 (2020)].

PACS numbers: 24.10.Cn, 71.20.Be, 71.10.Fd

Transition-metal dichalcogenides (TMDCs) [1, 2] are a class of materials of the typeMX$_2$, where M is a transition-metal atom (Mo, W, V, Hf, etc) and X is a chalcogen atom (S, Se, Te, etc). In recent decades, interest is grown rapidly in TMDCs due to their impressive electronic [1, 3–7], optical [2, 3] and mechanical properties [8], and the broad application to electronics [9–11], spintronics [12, 13], valleytronics [14, 15], optoelectronics [2, 16], and sensing [17]. When bulk TMDCs are thinned to monolayers, correlation effects become much more important than that in the bulk, because the three dimension Coulomb interaction is only screened in two dimensions, which results in a weak dielectric screening [18]. Many experiments have demonstrated the existence of strong electron-electron (e-e) interaction in monolayer TMDCs (ML-TMDCs), including interaction induced giant paramagnetic response in ML-MoS$_2$ [19], new photoluminescence peaks in ML-WX$_2$ ($X=\text{S, Se}$) [20, 21], enhanced valley magnetic response and quantum Hall states sequence transition in ML-WSe$_2$ [22, 23]. Optical susceptibility measurements of the molybdenum disulfide (MoS$_2$) monolayer in van der Waals heterostructure provided by Roch et al. show that e-e interactions, especially the intervalley exchange interaction, result in a first order phase transition from a spin unpolarized ground state to a spin polarized state in presence of an external magnetic field $\mathbf{B}$ [24–26].

In the photoluminescence spectrum, an abrupt change marks this first order phase transition when the trion peak ($X^-$) evolves into the Mahan exciton peak (Q) [25]. This first order phase transition attributes to the nonanalytic correction in the free energy [25, 27]. Without $\mathbf{B}$, the same abrupt change is still observed, which implies that a magnetic order transition occurs like the case of nonzero $\mathbf{B}$ [25]. However, the total magnetization is zero in the whole process, which seems to indicate that the transition of the magnetic order doesn’t occur. It is confusing. Roch et al. [25] proposed that the fluctuation between “puddles” of the spin up and spin down leads to the zero total magnetization at low electron density. However, there is no theoretical demonstration of the “puddles” (the degenerate spin polarized states). In previous theoretical studies [28, 29] intervalley e-e interaction was ignored and the spin-spin couplings in the intravalley and intervalley were not appreciated, which play a vital role, as shown by our results, in determining the properties of the ground state. We are motivated by the zero magnetic field experimental observations and the lacking of the theoretical explanation. Therefore, we focus on this case and try to understand the peculiar observations in experiments. In this paper, we study a model for ML-MoS$_2$ with including the intravalley and intervalley Coulomb interaction, based on the low-energy noninteracting Hamiltonian derived in previous studies [13] and develop a self-consistent mean field method (SCFM), emphasizing the effective intervalley spin-spin couplings.

It is found that the ground state is composed of two degenerated spin-polarized states at a certain electron density, giving rise to a zero total magnetization. By tuning the electron density via the chemical potential, a first order phase transition occurs between the unpolarized state to the spin-polarized states, which is consistent with the experiment [25].

Fig. 1 shows the crystal structure of ML-MoS$_2$ and its first Brillouin zone (BZ) [30]. The minima of the conduction band are located at the corners ($K, K'$). For the
description of the noninteracting case, we use the effective Hamiltonian of ML-MoS$_2$ around the Dirac cones \cite{13, 31} as

$$\hat{H}_0 = at (\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z - \lambda_\tau \sigma_z - \frac{1}{2} \bar{s}_z,$$  \hspace{1cm} (1)

where $\tau = \pm 1$ are valley indexes for $\mathbf{K}$ and $\mathbf{K}'$ (see Fig. 1). The spin splitting caused by spin orbit coupling is $2\lambda$. $\sigma_\alpha \ (\alpha = x, y, z)$ are the Pauli matrices. $a$ is the lattice constant. $t$ is the hopping integral. $\Delta$ is the energy gap between the conduction band and valence band (when $\lambda = 0$). $\bar{s}_z$ is the $z$ component of the spin operator. For convenience, BZ is chosen as the diamond region in the following calculation (see Fig. 1 (b)). The energy eigenvalues of the Hamiltonian are \cite{32}

$$E_{n_\tau s} = \lambda \tau s/2 \pm \sqrt{(atk)^2 + (\Delta - \lambda \tau s)/2)^2},$$  \hspace{1cm} (2)

where $s = \pm 1$ are the spin indexes for spin up and down respectively. The up plus (bottom minus) sign in Eq. (2) denotes the conduction (valence) band [c (v)]. $k$ is the module of the wave vector. The corresponding eigenstates are denoted as $|n_\tau ks\rangle$, where $n = c$ or $v$.

The Coulomb interaction between electrons is

$$V(r_1 - r_2) = \frac{e^2}{4\pi\varepsilon_0} \frac{1}{|r_1 - r_2|},$$  \hspace{1cm} (3)

where $e$ is the elementary charge, $\varepsilon_0$ is the vacuum permittivity. It is secondly quantized in the $|n_\tau ks\rangle$ representation \cite{33}

$$\hat{V} = \frac{1}{N} \sum_{n_1n_2\tau_1\tau_2} \sum_{k_1k_2} \sum_{s_1s_2} V_{\text{int}} a_{k_1s_1}^{\tau_1\dagger} a_{k_2s_2}^{\tau_2\dagger} a_{k_2s_2}^{\tau_2} a_{k_1s_1}^{\tau_1\dagger},$$  \hspace{1cm} (4)

where $V_{\text{int}}$ denotes the strength of the e-e interaction and $N$ is the number of unit cells. $a_{k_\tau s}^{\tau\dagger}$ ($a_{k_\tau s}^{\tau}$) is the creation (annihilation) operator in $|n_\tau ks\rangle$ state. There are three kinds of e-e interaction: interaction between the conduction electrons, interaction between the valence electrons and the interaction between the conduction electrons and the valence electrons. Here, we only take the interaction between the conduction electrons into consideration and eliminate the letter $c$, which is used to mark the conduction band, in the following formulas for convenience. The strength of the e-e interaction in the conduction band is written as

$$V_{\text{int}} = \frac{1}{2N} \langle \tau\bar{k}_1s_1, \tau\bar{k}_2s_2|V(r_1 - r_2)|\tau\bar{k}_3s_3, \tau\bar{k}_3s_3\rangle,$$  \hspace{1cm} (5)

$\hat{V}$ in Eq. (A37) is thus expanded explicitly and approximated as

$$\hat{V} \approx \hat{V}_{\text{intra}} + \hat{V}_{\text{inter}},$$  \hspace{1cm} (6)

where

$$\hat{V}_{\text{intra}} = \frac{1}{N} \sum_{k_1k_2} \sum_{\tau s} U a_{k_1s_1}^{\tau\dagger} a_{k_2s_2}^{\tau\dagger} a_{k_2s_2}^{\tau} a_{k_1s_1}^{\tau\dagger},$$  \hspace{1cm} (7)

$$\hat{V}_{\text{inter}} = \frac{1}{N} \sum_{k_1k_2} \sum_{\tau s_1s_2} U' a_{k_1s_1}^{\tau\dagger} a_{k_2s_2}^{\tau\dagger} a_{k_1s_1}^{\tau} a_{k_2s_2}^{\tau\dagger},$$  \hspace{1cm} (8)

$\hat{V}_{\text{intra}}$ and $\hat{V}_{\text{inter}}$ denote the intravalley and intervalley e-e interaction respectively. $U$ and $U'$ are the strengths of the corresponding e-e interaction,

$$U = \frac{1}{2N} \langle \tau\bar{k}_1, \tau\bar{k}_2|V(r_1 - r_2)|\tau\bar{k}_1, \tau\bar{k}_2\rangle,$$  \hspace{1cm} (9)

$$U' = \frac{1}{2N} \langle \tau\bar{k}_1, \tau\bar{k}_2|V(r_1 - r_2)|\tau\bar{k}_2, \tau\bar{k}_1\rangle.$$  \hspace{1cm} (10)

$\bar{s} \ (s)$ represents the opposite valley (spin) of $s \ (\bar{s})$, $k \ (\bar{k})$ indicates the relative wave vector with respect to the minimum of $\tau \ (\bar{\tau})$ valley. Quantitatively, it has been estimated in the static screening limit that due to the small Bohr radius the intervalley e-e interaction is comparable to the intravalley interaction even at high electron density \cite{26}. Hence, it is necessary to take the intervalley e-e interaction into consideration when one deals with the Coulomb interaction in TMDCs \cite{26}. For the purpose of a qualitative discussion, $U$ and $U'$ are regarded as constants. Details of the above approximation are shown in appendix A.

We apply the mean field approximation \cite{33} to $\hat{V}_{\text{intra}}$ and $\hat{V}_{\text{inter}}$ respectively. As for $\hat{V}_{\text{intra}}$, it reads

$$\hat{V}_{\text{intra}} \approx \sum_{k_\tau s} \left( U_{\tau s} a_{k_\tau s}^{\tau\dagger} a_{k_\tau s}^{\tau\dagger} + \bar{U}_{\tau s} a_{k_\tau s}^{\tau\dagger} a_{k_\tau s}^{\tau\dagger} \right),$$  \hspace{1cm} (11)

where

$$U_{\tau s} = \frac{2}{N} \sum_{k} U\langle a_{k_\tau s} \rangle, \quad \bar{U}_{\tau s} = \frac{2}{N} \sum_{k} U\langle n_{k_\tau s}^\tau \rangle,$$  \hspace{1cm} (12)

$n_{k_\tau s} = a_{k_\tau s}^{\tau\dagger} a_{k_\tau s}^{\tau}$ is the particle number operator. In this paper, we merely consider the zero temperature case. Therefore, $\langle \cdots \rangle$ means the ground state average. In
terms of the spin operators, \( S_{\tau k}^z = \frac{1}{2} (n_{k\uparrow}^\tau - n_{k\downarrow}^\tau) \),
\( S_{\tau k}^+ = a_{k\uparrow}^\dagger a_{k\downarrow} \) and \( S_{\tau k}^- = a_{k\downarrow} a_{k\uparrow}^\dagger \), \( \hat{V}_{\text{inter}} \) is rewritten as
\[
\hat{V}_{\text{inter}} = -\frac{U'}{N} \sum_{k, k'} \left( n_{k\uparrow} n_{k'\downarrow}^\ast + 4 S_{\tau k1} \cdot S_{\tau k2} \right),
\]
where \( n_{k} = \sum_{s} n_{ks} \), \( S \) is the spin operator and
\[
S_{\tau k1} \cdot S_{\tau k2} = S_{\tau k1}^z S_{\tau k2}^z + \frac{1}{2} \left( S_{\tau k1}^+ S_{\tau k2}^- + S_{\tau k1}^- S_{\tau k2}^+ \right).
\]
In Eq. (13), the first and second term give the intervalley density-density interaction and the intervalley spin-spin coupling. We apply mean field approximation to Eq. (13) and obtain
\[
\hat{V}_{\text{inter}}^{\text{MF}} \approx -\sum_{k} \left( U' n_{k}^{\tau} + U' n_{k}^{\bar{\tau}} + M_{\tau} S_{\tau k}^z + M_{\bar{\tau}} S_{\bar{\tau} k}^z \right),
\]
where
\[
U'_{\tau(k)} = \frac{1}{N} \sum_{k(k')} U' (n_{k(k')}^{\tau}), \quad M_{\tau} = \frac{4}{N} \sum_{k(k')} U' (S_{\tau(k(k))}^z).
\]
In Eq. (13), the direction of \( S \) is chosen as the \( z \)-axis. By defining
\[
\chi_{\tau \bar{\tau}} = U'_{\tau} + \frac{1}{2} s M_{\tau}, \quad \chi_{\bar{\tau} \tau} = U'_{\bar{\tau}} + \frac{1}{2} s M_{\bar{\tau}},
\]
where \( s = \pm 1 \) for states with spins parallel and antiparallel to the \( S \), and we obtain
\[
\hat{V}_{\text{inter}}^{\text{MF}} \approx -\sum_{k} \left( \chi_{\tau k} a_{k\uparrow}^\dagger a_{k\downarrow} + \chi_{\bar{\tau} k} a_{k\downarrow}^\dagger a_{k\uparrow} \right).
\]
Therefore, the mean field approximation of the interaction, i.e., \( \hat{V}_{\text{MF}} = \hat{V}_{\text{inter}}^{\text{MF}} \), is
\[
\hat{V}_{\text{MF}} = \sum_{k} \left( F_{\tau k} a_{k\uparrow}^\dagger a_{k\downarrow} + F_{\bar{\tau} k} a_{k\downarrow}^\dagger a_{k\uparrow} \right).
\]
where \( F_{\tau k} \) and \( F_{\bar{\tau} k} \) are the effective mean fields, which read
\[
F_{\tau k} = U_{\tau k} - \chi_{\tau k}, \quad F_{\bar{\tau} k} = U_{\bar{\tau} k} - \chi_{\bar{\tau} k}.
\]
The total mean field Hamiltonian reads
\[
H_{\text{total}}^{\text{MF}} = \sum_{k} \left( E_{\tau k} (a_{k\uparrow}^\dagger a_{k\downarrow} + E_{\bar{\tau} k} (a_{k\downarrow}^\dagger a_{k\uparrow}) \right),
\]
where the energy spectrum
\[
E_{\tau k}(\tau(k)) = E_{\tau k}(\tau(k)) + F_{\tau(k)} - \mu,
\]
where \( \mu \) is the chemical potential. In above mean field approximation we omit the constant terms, which do not affect our general discussions and qualitative conclusions. The constant terms neglected in the calculations merely shift all energy bands simultaneously. This leads us a zero-energy redefinition. This shift cannot affect the determination of the solutions which are determined by the parameters that are not entangled with the absolute energies but the relative energy with respect to the zero-energy. It is easy then to calculate the free energy
\[
E_{\text{free}} = \frac{1}{N} \sum_{k} \int E_{\tau k}(k) dk.
\]
The detailed calculations of \( E_{\text{free}} \) can be found in appendix C. In order to calculate the effective mean fields, averages \( \langle n_{ks} \rangle \) need to be calculated. We thus introduce
\[
\tilde{n}_{\tau} = \frac{1}{N} \sum_{k} \langle n_{ks} \rangle.
\]
The total electron number per unit cell at \( \tau \) valley is \( \tilde{n}_{\tau} = \sum \tilde{n}_{\tau}^\dagger \), lying in a domain of \([0, 1]\). It is convenient for the following discussion to define the valley magnetization as
\[
m_{\tau} = \tilde{n}_{\tau} - \tilde{n}_{\bar{\tau}}, \quad m_{\bar{\tau}} = \tilde{n}_{\bar{\tau}} - \tilde{n}_{\bar{\tau}}^\dagger,
\]
which indicate the valley spin polarization. The total magnetization is then \( m = m_{\tau} + m_{\bar{\tau}} \). When the ground state is spin polarized, the total magnetization \( m \neq 0 \). In contrast, \( m = 0 \). In terms of \( \tilde{n}_{\tau} \) and \( m_{\tau} \), the mean field is rewritten as
\[
F_{\tau k} = U (\tilde{n}_{\tau(k)} + s m_{\tau(k)}) - U' (\tilde{n}_{\tau(k)} + s m_{\tau(k)}).
\]
The gap of the spin splitting of the conduction band is readily obtained
\[
\Delta_{E_{\tau}}(k) = E_{\tau(k)} - E_{\tau(k)}(k) + \Delta_{F},
\]
where \( \Delta_{F} = -2 U m_{\tau} - 2 U' m_{\tau} \), which shows the influence of the e-e interaction on the spin splitting of the
conduction band, and indicates the renormalization of the conduction band minimum (CBM). The renormalized position of CBM is self-consistently calculated. Parameters \( \tilde{n}_\tau, m_\tau, \tilde{m}_\tau \) constitute a four dimension parameter space. Any point in the space is denoted as a vector \((\tilde{n}_\tau, m_\tau, \tilde{m}_\tau, m_\tau)\). At this stage, we have obtained all of mean field equations, which are solved numerically and self-consistently. The procedure of the numerical calculation is as following. At first, we give a set of values for \( \tilde{n}_\tau, m_\tau, \tilde{m}_\tau \), and \( m_\tau \), which corresponds to a vector \( \mathbf{P}_{\text{given}} \) in the parameter space. Then, the effective mean field \( \mathbb{F}_{\tau s} \) is obtained by substituting \( \tilde{n}_\tau, m_\tau, \tilde{m}_\tau \), and \( m_\tau \) into Eq. 25. Utilizing Eq. 21, we get the energy spectrum. Finally, we calculate \( \Delta_i \) by Eq. (22) and update \( \tilde{n}_\tau, m_\tau, \tilde{m}_\tau \), and \( m_\tau \) via Eq. (C12) and Eq. (24). Note that parameter \( \tilde{n}_\tau \) should be calculated via an integral over the momentum space. And other parameters are not generated from integrals but from \( \tilde{n}_\tau \). At this stage, we have obtained \( \Delta_i \) and \( \tilde{n}_\tau \) directly (see Eq. (24)). \( \tilde{n}_\tau \) is determined by the relative position of the energy with respect to the Fermi level (or the chemical potential) (see appendix C). Hence, we neglect constant terms in the mean-field process which shift all energy bands equally and have no effect on the integral of \( \tilde{n}_\tau \) and the self-consistent process. The updated \((\tilde{n}_\tau, m_\tau, \tilde{m}_\tau)\) corresponds to a new point in the parameter space, denoted by a vector \( \mathbf{P}_{\text{update}} \). We define the distance of the two points as the deviation

\[
\delta = |\mathbf{P}_{\text{given}} - \mathbf{P}_{\text{update}}|.
\]

For a given point in the parameter space, if it is a solution of the set of mean field equations (MFEs), then \( \delta \) is zero. We thus scan the entire parameters space and try to find the parameter vectors where \( \delta \) converges to zero. And we define these parameter vectors as the solutions.

It is found that the solution of MFEs is not unique (see Fig. 2). And the solutions are characterized by converged parameter vectors in the parameter space. In the numerical calculation, we grid the definition domain of each parameters into \( N \) subintervals, that is, the parameter space is grided into \( N^4 \) subspaces. As \( N \) goes to infinite, the parameter space is ergodic exactly. In practice, we take a finite \( N \), and \( \delta \) is kept at the order \( 10^{-6} \). In this paper, we take \( a = 3.193 \, \text{Å}, \ t = 1.1 \, \text{eV}, \Delta = 1.66 \, \text{eV}, \) and \( 2\lambda = 0.15 \, \text{eV} \), which are the fitting results to the ab initio calculation [13]. The intravalley Coulomb interaction \( U \) is usually unknown in TMDCs. According to the discussion of R. Roldán et al. [34], electronic states in the neighbor region of \( \mathbf{K} \) and \( \mathbf{K}' \) points are characterized by the 4d orbitals of Mo atoms. The order of magnitude of \( U_{dd} \) is approximated by the ionization energy of Mo atom. According to previous investigations of MoS\(_2\) [34, 35], one usually takes \( U_{dd} \approx 2.0 \sim 4.0 \, \text{eV} \). We compare our definition of the interacting Hamiltonian with that of Rostami [35], we find \( U = U_{dd}/2 \). Therefore, the intravalley Coulomb interaction is about \( U \approx 1.0 \sim 2.0 \, \text{eV} \). Basing on the above consideration, we take \( U = 1.0 \, \text{eV} \) and find the result is reasonable, when \( U \) is combined with other parameter values.

Fig. 2 shows the evolution of \( \Delta_i \) and \( \tilde{n}_\tau \) with \( \tilde{n}_\tau, m_\tau, \tilde{m}_\tau, \) and \( m_\tau \) in (a)-(d), respectively (\( N = 150 \)). We obtain three solutions: one solution \( L_0 \) and two degenerated solutions \( L_\pm \) (with the same free energy). \( \Delta_i \) is not the total free energy because the constant terms have been neglected in the calculation (see Eq. (B1) and Eq. (B6)). As for different mean field solutions \( (L_0 \text{ and } L_\pm) \), the values of the neglected constant are different. In Fig.2, we show \( \Delta_i \) of \( L_0 \) and \( L_\pm \) on the same energy scale for convenience without the meaning of comparison. \( L_\pm \) are new solutions, which haven’t been obtained previously due to the ignoring of the intervalley Coulomb interaction [28, 29]. In Fig. 2(a) and (c), \( \tilde{n}_\tau(L_+) \) and \( \tilde{n}_\tau(L_-) \) are very close (the same to the solution of \( L_- \)). At the numerical precision \( \delta \approx 10^{-6} \), we obtain the difference \( \tilde{n}_\tau(L_+) - \tilde{n}_\tau(L_-) \) is not zero but about the order of \( 10^{-4} \) indicating a slight valley polarization. However, \( \tilde{n}_\tau = \tilde{n}_\times \) for \( L_0 \) (see Fig. 2(a) and (c)). As for \( L_0 \), the states of \( \tau \) and \( \bar{\tau} \)-valley can be spin polarized but in opposite directions, which contributes a zero net magnetization (Fig. 2(b) and (d)). In contrast, for solutions \( L_+ \) and \( L_- \), spin polarization for both valleys can be induced as well but in the same direction, leading to a net magnetization for each solution. Because \( L_\pm \) are two degenerated solutions, the spin-polarized states (composed of two valleys) from \( L_+ \) and \( L_- \) are aligned opposite, giving rise to a zero net magnetization since the state of entire system is randomly composed of the states of \( L_+ \) and \( L_- \). [25]. We further speculate that the states of \( L_\pm \) may manifest themselves by forming spin-polarized “domains” in real materials. And globally there is no net magnetization without introducing an external magnetic field.

We firstly discuss \( U' = 0 \) case, and the effective mean field becomes \( \mathbb{F}_{\tau s} = \frac{U}{2} \sum_k \langle n^{\tau}_{k\delta} \rangle \). We derive the solution \( L_0 \) that satisfies \( \sum_k \langle n^{\tau}_{k\delta} \rangle = \sum_k \langle n^{\bar{\tau}}_{k\delta} \rangle \), and \( \sum_k \langle n^{\tau}_{k\delta} \rangle = \sum_k \langle n^{\bar{\tau}}_{k\delta} \rangle / U \). These indicate that the spin splitting of the conduction band at \( \tau \) and \( \bar{\tau} \) is inverted.

![Fig. 3](image_url)

**FIG. 3.** (Color online) (a) The dependence of \( m_\tau, \tilde{m}_\tau \) and \( m \) on \( \mu \). \( U = 1.0 \, \text{eV}, U' = 0 \, \text{eV} \). (b) and (c) The energy spectrum \( E_{\text{corr}}(k) \) along \( k_x \) direction. \( U = 1.0 \, \text{eV}, U' = 0 \, \text{eV} \) and \( \mu = 1.7 \, \text{eV} \). The horizontal line shows the Fermi surface.
due to time-reversal symmetry (TRS) (see Fig. 3(b) and (c)) \[13\]. It can be seen from energy gap in Eq. (26) at the minimum of the conduction band \(k = 0\), \(\Delta_{\pm}^0(0) = -2Um_\tau\). From Fig. 3(a), we find that \(m_\tau = -m_\sigma\), and then \(\Delta_{\pm}^0(0)\) always takes the opposite values (see Fig. 3(b) and (c)). We only derive the \(L_0\) solution in this case, which means an unpolarized state in absence of intervalley interaction.

In general, the intervalley e-e interaction is comparable to the intravalley interaction even at high electron density, due to a small Bohr radius \(a_B \approx 0.5\) nm \[26\]. In this case, all of \(\tilde{n}_\sigma, m_\tau, \tilde{n}_\tau\), and \(m_\sigma\) appear in the mean field \(F_{\tau\bar{n}}\) (Eq. (25)), i.e. carriers in the two valleys are interacted with each other. Fig. 4(a) shows the dependence of \(m\) on \(\mu\) at various \(U'\) for \(L_+\) state. The main feature of the \(L_+\) state is that there is a region of gate voltage (characterized by chemical potential \(\mu\)) in which a net magnetization is developed and characterized by a finite \(m\). However, the global net magnetization is zero due to the superposition of the \(L_\pm\) states. We may call it as a ferromagnetic (FM) state. Out of this region, \(m = 0\) means a paramagnetic (PM) state. Therefore, there are two borders between the FM and PM state located at a lower and a higher \(\mu\), which indicate a PM-FM phase transition and vice versa. However, three kinds of transitions are found. The first one is shown for \(U' = 0.2\) eV, where \(m\) grows and disappears with \(\mu\) continuously, indicating a second-order phase transition at two borders; the second one is shown for \(U' = 0.4, 0.6\), and 0.8 eV, where the PM-FM transitions are emergent discontinuously, which indicate a first-order phase transition and consistent with the experimental observations \[25\]. The third one \((U' = 0.1\) eV\) shows a second-order and a first-order at the lower and higher \(\mu\), respectively. As a theoretical investigation, we study the effects due to various parameters to cover most possibilities. The validity of the parameters should get supports from experimental observations or other ways. Our results show that when \(U' \geq 0.4\) eV, the phase transition is clearly of first order, which agrees with the experimental results \[25\]. This is also consistent with the previous prediction that the intervalley interaction is comparable with the intravalley interaction even at high electron density \[26\]. We therefore deduce that realistic intervalley Coulomb interaction should be in this range. The complicated transition behaviors exhibited in other parameter ranges might not be a reality. It is still lack of an intuitive picture for appearance of such a complicated case.

To understand the existence of the FM state, we show band structures in Figs. 4(b)-(m) and the relative positions of Fermi level to CBM. The electron Coulomb interaction renormalizes the band structures (or the position of CBM). For different \(U'\), the relative position of the Fermi level (or chemical potential) to the CBM is different. The PM states at small \(\mu\) can be understood because the Fermi level does not pass through any bands (Figs. 4(b), (c), (h) and (i)). For the PM states at large \(\mu\) (Figs. 4(f), (g), (l) and (m)), the Fermi level deeply lies in all four bands where the e-e interaction may be weak due to a high electron density, and the spin splitting due to the e-e interaction is insignificant. In contrast to these two cases, Figs. 4(d), (e), (j) and (k) show the band structures for the FM states. It is noted that the spin splitting of bands is obviously observed and the Fermi level is not deeply lying in the conduction bands, but lies just around the bottom of some bands. This result matches our intuitive picture that the electron Coulomb interaction should be more important when the Fermi level is close to the CBM. \[24\] For Figs. 4(d) and (e), it seems a “normal” FM state in which the Fermi level is not far away from the bottom of four bands. However, for Figs. 4(j) and (k), the Fermi level is deeply in the spin-up bands but shallowly lies in the spin-down bands. It might be this difference leading to a different transition order between the PM and FM existing at lower and higher \(\mu\), respectively. It is obvious that the relative position of the Fermi level and the CBM is quite crucial for existence of the FM states. And this relative position is altered dramatically by including the electron Coulomb interaction and can not qualitatively predicted by thinking about the picture of non-interaction case.

The complicate behaviors of the transition induced by \(\mu\) may rest themselves into a fact that the energy bands are altered in the self-consistent MFEs. Comparable to the experiments, it seems that the first-order phase transitions at the two borders may be consistent with experimental observations \[25\]. If this is the case, we can

FIG. 4. (Color online) (a) The dependence of \(m\) on \(\mu\) at various \(U'\). With the increase of \(\mu\), the solution of MFEs changes between \(L_0\) and \(L_\pm\). When the solutions are \(L_\pm\), we merely chose \(L_+\) and calculate \(m\) of \(L_+\), because \(L_+\) and \(L_-\) are degenerate. (b)-(m) The spin splitting of the conduction band along the \(k_z\) direction. The horizontal lines represent the Fermi level. \(U = 1.0\) eV.
deduce that the $U'$ may be in the range of 0.4-0.8 eV. This energy scale may be converted to a length scale which corresponds to a Coulomb length for $U'$ and a size of the so-called “puddle” in experiments, which is in 1-2 nm. This can be tested in experiments although this size is said to be small but not given in experiments. So far, we know that the polarized “puddles” resemble the domains in usual ferromagnets. In zero-B case, the polarizations of these “puddles” may be randomly distributed giving rise to a zero net magnetization. We may speculate that the polarizations of “puddles” may be aligned into one direction when applying a nonzero B, which bring us a net magnetization. This scenario is consistent with the experimental observation [24]. A further measurement on this size can demonstrate our theory clearly. We should emphasize that an FM state can be induced by tuning gate voltage due to finite $U'$. This reflects the important role of intervalley Coulomb interaction. The FM state can be derived only in the presence of intervalley Coulomb interaction. In the absence of the Coulomb interaction, Eq.(2) shows that the ground state is both valley and spin degenerate at k=0. The conduction band at $\tau$ and $\bar{\tau}$ is inverted as the requirement of TRS. However, in the presence of the Coulomb interaction, it is found that the valley and spin degeneracy are lifted (Fig. 4(d), (e), (j), (k)). Note that the energy difference caused by spin orbital coupling is $2\lambda$. $\sigma_\alpha (\alpha = x, y, z)$ is the Pauli matrices. $a$ is the lattice constant. $t$ is the hopping integral. $\Delta$ is the energy gap between the conduction band and the valence band (when $\lambda = 0$). $\hat{s}_z$ is the z component of the spin operator. For convenience, we choose a diamond Brillouin zone (BZ) in the following calculation (see Fig. 6). We explicitly write

$$\hat{s}_z = \begin{pmatrix} s_{z_1} & 0 \\ 0 & s_{z_2} \end{pmatrix}, \quad \tau = \begin{pmatrix} \tau_1 & 0 \\ 0 & \tau_2 \end{pmatrix},$$

where $s_{z_1} = 1$ and $s_{z_2} = -1$ represent spin up and spin down respectively, $\tau_1 = 1$ and $\tau_2 = -1$ indicate the two valleys located at K and $K'$. We perform direct product for the valley, spin and band (conduction band and valence band) index freedom in the Hamiltonian. $\hat{H}_0$ is rewritten as

$$\hat{H}_0 = at(\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z - \lambda \tau \frac{\sigma_z - 1}{2} \hat{s}_z,$$

where $\tau = \pm 1$ is the valley index. The spin splitting caused by spin orbital coupling is $2\lambda$. $\sigma_\alpha (\alpha = x, y, z)$ is the Pauli matrices. $a$ is the lattice constant. $t$ is the hopping integral. $\Delta$ is the energy gap between the conduction band and the valence band (when $\lambda = 0$). $\hat{s}_z$ is the z component of the spin operator. For convenience, we choose a diamond Brillouin zone (BZ) in the following calculation (see Fig. 6). We explicitly write

$$\hat{s}_z = \begin{pmatrix} s_{z_1} & 0 \\ 0 & s_{z_2} \end{pmatrix}, \quad \tau = \begin{pmatrix} \tau_1 & 0 \\ 0 & \tau_2 \end{pmatrix},$$

where $s_{z_1} = 1$ and $s_{z_2} = -1$ represent spin up and spin down respectively, $\tau_1 = 1$ and $\tau_2 = -1$ indicate the two valleys located at K and $K'$. We perform direct product for the valley, spin and band (conduction band and valence band) index freedom in the Hamiltonian. $\hat{H}_0$ is rewritten as

$$\hat{H}_0 = at(\tau \otimes k_x \sigma_x \otimes 1 + 1 \otimes k_y \sigma_y \otimes 1) + 1 \otimes \frac{\Delta}{2} \sigma_z \otimes 1 - \lambda \tau \otimes \frac{\sigma_z - 1}{2} \otimes \hat{s}_z,$$

where 1 is the identity matrix, and $\otimes$ denotes direct product. It is obvious that $\hat{H}_0$ is a 8 $\times$ 8 matrix. Substituting the Pauli matrix,

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$

into Eq.(A3), one obtains

$$\hat{H}_0 = \begin{pmatrix} a & 0 \\ 0 & \beta \end{pmatrix}.$$

$\hat{H}_0$ is a block matrix, where 0, $\alpha$ and $\beta$ are 4 $\times$ 4 matrices. 0 is a zero matrix,

$$\alpha = \begin{pmatrix} \frac{\Delta}{2} & 0 & \alpha_- & 0 \\ 0 & \frac{\Delta}{2} & 0 & \alpha_- \\ \alpha_+ & 0 & -\frac{\Delta}{2} + \lambda \tau_1 s_{z_1} & 0 \\ 0 & \alpha_+ & 0 & -\frac{\Delta}{2} + \lambda \tau_1 s_{z_2} \end{pmatrix},$$

where $\alpha_\pm = at(\tau_1 k_x \pm ik_y)$, and

$$\beta = \begin{pmatrix} \frac{\Delta}{2} & 0 & \beta_- & 0 \\ 0 & \frac{\Delta}{2} & 0 & \beta_- \\ \beta_+ & 0 & -\frac{\Delta}{2} + \lambda \tau_2 s_{z_1} & 0 \\ 0 & \beta_+ & 0 & -\frac{\Delta}{2} + \lambda \tau_2 s_{z_2} \end{pmatrix},$$

Appendix A: Model

1. Solve noninteracting Hamiltonian

According to the work reported by Xiao Di et al. [13], the effective Hamiltonian of ML-MoS₂ around Dirac cones without Coulomb interaction is

$$\hat{H}_0 = at(\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z - \lambda \tau \frac{\sigma_z - 1}{2} \hat{s}_z,$$
FIG. 5. (Color online) Energy spectrum along \( k_x \) direction at \( \tau_1 \) (Left) and \( \tau_2 \) (Right) valley. \( a = 3.193 \text{ Å}, t = 1.1 \text{ eV}, \Delta = 1.66 \text{ eV}, \) and \( 2\lambda = 0.15 \text{ eV}. \)

where \( \beta_{\pm} = at(\tau_2 k_x \pm ik_y) \). It is easy to diagonalize \( \alpha \) matrix. The energy eigenvalues for \( \tau_1 \) valley reads

\[
E^{(1)}_{\tau_1 s_1} = \frac{\lambda \tau_1 s_{z_1}}{2} - \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_1 s_{z_1}}{2}\right)^2},
\]

\[
E^{(2)}_{\tau_1 s_1} = \frac{\lambda \tau_1 s_{z_1}}{2} + \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_1 s_{z_1}}{2}\right)^2},
\]

\[
E^{(3)}_{\tau_1 s_2} = \frac{\lambda \tau_1 s_{z_2}}{2} - \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_1 s_{z_2}}{2}\right)^2},
\]

\[
E^{(4)}_{\tau_1 s_2} = \frac{\lambda \tau_1 s_{z_2}}{2} + \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_1 s_{z_2}}{2}\right)^2}.
\]

The corresponding eigenvectors are

\[
u^{(1)}_{\tau_1 s_1} = N^{(1)}_{\tau_1 s_1} \left(\frac{\Delta - 2E^{(2)}_{\tau_1 s_1}}{2at(\tau_1 k_x + ik_y)}, 0, 1, 0\right)^T,
\]

\[
u^{(2)}_{\tau_1 s_1} = N^{(2)}_{\tau_1 s_1} \left(\frac{\Delta - 2E^{(1)}_{\tau_1 s_1}}{2at(\tau_1 k_x + ik_y)}, 0, 1, 0\right)^T,
\]

\[
u^{(3)}_{\tau_1 s_2} = N^{(3)}_{\tau_1 s_2} \left(0, \frac{\Delta - 2E^{(4)}_{\tau_1 s_2}}{2at(\tau_1 k_x + ik_y)}\right)^T,
\]

\[
u^{(4)}_{\tau_1 s_2} = N^{(4)}_{\tau_1 s_2} \left(0, \frac{\Delta - 2E^{(3)}_{\tau_1 s_2}}{2at(\tau_1 k_x + ik_y)}\right)^T.
\]

where the eigenvectors are normalized by

\[
N^{(1)}_{\tau_1 s_1} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(2)}_{\tau_1 s_1})^2 + 4(at)^2}},
\]

\[
N^{(2)}_{\tau_1 s_1} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(1)}_{\tau_1 s_1})^2 + 4(at)^2}},
\]

\[
N^{(3)}_{\tau_1 s_2} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(4)}_{\tau_1 s_2})^2 + 4(at)^2}},
\]

\[
N^{(4)}_{\tau_1 s_2} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(3)}_{\tau_1 s_2})^2 + 4(at)^2}}.
\]

In the same way, we diagonalize \( \beta \) matrix obtaining the energy eigenvalues at \( \tau_2 \) valley

\[
E^{(5)}_{\tau_2 s_1} = \frac{\lambda \tau_2 s_{z_1}}{2} - \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_2 s_{z_1}}{2}\right)^2},
\]

\[
E^{(6)}_{\tau_2 s_1} = \frac{\lambda \tau_2 s_{z_1}}{2} + \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_2 s_{z_1}}{2}\right)^2},
\]

\[
E^{(7)}_{\tau_2 s_2} = \frac{\lambda \tau_2 s_{z_2}}{2} - \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_2 s_{z_2}}{2}\right)^2},
\]

\[
E^{(8)}_{\tau_2 s_2} = \frac{\lambda \tau_2 s_{z_2}}{2} + \sqrt{(at)^2 + \left(\frac{\Delta - \lambda \tau_2 s_{z_2}}{2}\right)^2}.
\]

The eigenvectors are

\[
u^{(5)}_{\tau_2 s_1} = N^{(5)}_{\tau_2 s_1} \left(0, \frac{\Delta - 2E^{(6)}_{\tau_2 s_1}}{2at(\tau_2 k_x + ik_y)}\right)^T,
\]

\[
u^{(6)}_{\tau_2 s_1} = N^{(6)}_{\tau_2 s_1} \left(0, \frac{\Delta - 2E^{(5)}_{\tau_2 s_1}}{2at(\tau_2 k_x + ik_y)}\right)^T,
\]

\[
u^{(7)}_{\tau_2 s_2} = N^{(7)}_{\tau_2 s_2} \left(0, \frac{\Delta - 2E^{(8)}_{\tau_2 s_2}}{2at(\tau_2 k_x + ik_y)}\right)^T,
\]

\[
u^{(8)}_{\tau_2 s_2} = N^{(8)}_{\tau_2 s_2} \left(0, \frac{\Delta - 2E^{(7)}_{\tau_2 s_2}}{2at(\tau_2 k_x + ik_y)}\right)^T.
\]

where

\[
N^{(5)}_{\tau_2 s_1} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(6)}_{\tau_2 s_1})^2 + 4(at)^2}},
\]

\[
N^{(6)}_{\tau_2 s_1} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(5)}_{\tau_2 s_1})^2 + 4(at)^2}},
\]

\[
N^{(7)}_{\tau_2 s_2} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(8)}_{\tau_2 s_2})^2 + 4(at)^2}},
\]

\[
N^{(8)}_{\tau_2 s_2} = \sqrt{\frac{4(at)^2}{(\Delta - 2E^{(7)}_{\tau_2 s_2})^2 + 4(at)^2}}.
\]
Energy eigenvalues are written compactly as [32]

$$E_{n\tau s} = \frac{1}{2} \lambda \tau s \pm \sqrt{(at)^2 + \left(\frac{\lambda \tau s}{2}\right)^2},$$

(A32)

where the spin index $s = s_{\uparrow} \text{ or } s_{\downarrow}$. $E_{n\tau s}$ is shown in Fig. 5. The up plus sign denotes the conduction band $(c)$. The bottom minus sign denotes the valence band $(v)$. $n$ is the band index (conduction band $n = c$, valence band $n = v$). $k$ is the module of the wave vector, $k = \sqrt{k_x^2 + k_y^2}$. The valley index $\tau = \tau_1 \text{ or } \tau_2$. The corresponding eigenstate is a superposition state of the bases [13] with the coefficients defined by the eigenvectors, which is denoted as $|n\tau k s\rangle = \psi_{n\tau s}(k, r)$.

2. Coulomb interaction

Electron-electron (e-e) interactions have significant effects on the physical properties of monolayer materials [18]. As early as in 1979, Keldysh investigated Coulomb interaction in thin semiconductor and semimetal films, and gave an effective Coulomb interaction, which is expressed by the Neumann and Struve functions [37]. In this paper, we focused on a qualitative discussion. Therefore, we take the usual bare Coulomb interaction, instead of the complicated potential given by Keldysh. The bare Coulomb interaction is

$$V(r_1 - r_2) = \frac{e^2}{4\pi \varepsilon_0 |r_1 - r_2|},$$

(A33)

where $e$ is the elementary charge, $\varepsilon_0$ is the vacuum permittivity. It is obvious that in terms of field operators the Coulomb interaction is written as [33]

$$\hat{V}(r_1 - r_2) = \frac{e^2}{4\pi \varepsilon_0} \sum_{s_1, s_2} \int dr_1 dr_2 V(r_1 - r_2) \psi_{s_1}^\dagger(r_1) \psi_{s_2}^\dagger(r_2) \psi_{s_2}(r_2) \psi_{s_1}(r_1).$$

(A34)

We take the transformation

$$\psi_s^\dagger(r) = \frac{1}{\sqrt{N}} \sum_{n\tau k s} \psi_{n\tau s}(k, r) a_{n\tau k s}^\dagger,$$

(A35)

$$\psi_s(r) = \frac{1}{\sqrt{N}} \sum_{n\tau k s} \psi_{n\tau s}(k, r) a_{n\tau k s}^\dagger.$$  

(A36)

It is secondly quantized in the $|n\tau k s\rangle$ representation [33]

$$\hat{V} = \frac{1}{N} \sum_{s_1, s_2} \sum_{k_1, k_2} \sum_{\tau_1, \tau_2} \sum_{\tau_3, \tau_4} \sum_{\tau_5, \tau_6} \sum_{\tau_7, \tau_8} V_{\text{int}} a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1} a_{n_5 \tau_5 s_1} a_{n_6 \tau_6 s_1} a_{n_7 \tau_7 s_1} a_{n_8 \tau_8 s_1},$$

(A37)

where $V_{\text{int}}$ denotes the strength of the e-e interaction and $N$ is the number of the unit cell. $a_{n\tau s}^\dagger$ ($a_{n\tau s}^\dagger$) is the creation (annihilation) operator at $|n\tau k s\rangle$ state. Because the valence band is fully filled, we only consider the e-e interaction in the conduction band, i.e. $n = c$. In the following derivation, the superscript $c$ is omitted. We take the summation of $\tau_2, \tau_3$ and $\tau_4$ in Eq. (A37), obtaining

$$\hat{V} = \frac{1}{N} \sum_{\tau_1} \sum_{k_1, k_2} \sum_{\tau_3} \sum_{\tau_4} \sum_{i=1} V_{\text{int}}^i T_i,$$

(A38)

where $T_i$ reads

$$T_1 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A39)

$$T_2 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A40)

$$T_3 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A41)

$$T_4 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A42)

$$T_5 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A43)

$$T_6 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A44)

$$T_7 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1},$$

(A45)

$$T_8 = a_{n_1 \tau_1 s_1}^\dagger a_{n_2 \tau_2 s_2}^\dagger a_{n_3 \tau_3 s_2} a_{n_4 \tau_4 s_1}.$$  

(A46)

It is obvious that $T_1$ gives the intravalley e-e interaction. $T_2, T_3, T_4, T_5$ and $T_8$, describe the electron transformation from one valley to the other, which are not considered in this paper. $T_7$ shows the intravalley transformation of electrons without exchanges of spin, which is also neglected. $T_6$ gives the intervalley spin exchange coupling. The interaction induced magnetic order transition of ground state is attributed to this term, which is taken into consideration carefully. The strength of the interaction corresponding to $T_1$ reads

$$V_{\text{int}}^{(1)} = \frac{1}{2N} \langle \tau k_1, \tau k_2 | V(r_1 - r_2) | \tau k_4, \tau k_3 \rangle,$$

(A47)

$$V_{\text{int}}^{(2)} = \frac{1}{2N} \langle \tau k_1, \tau k_2 | V(r_1 - r_2) | \bar{\tau} k_4, \tau k_3 \rangle,$$

(A48)

$$V_{\text{int}}^{(3)} = \frac{1}{2N} \langle \tau k_1, \tau k_2 | V(r_1 - r_2) | \tau k_4, \bar{\tau} k_3 \rangle,$$

(A49)

$$V_{\text{int}}^{(4)} = \frac{1}{2N} \langle \tau k_1, \tau k_2 | V(r_1 - r_2) | \bar{\tau} k_4, \bar{\tau} k_3 \rangle,$$

(A50)

$$V_{\text{int}}^{(5)} = \frac{1}{2N} \langle \tau k_1, \bar{\tau} k_2 | V(r_1 - r_2) | \tau k_4, \tau k_3 \rangle,$$

(A51)

$$V_{\text{int}}^{(6)} = \frac{1}{2N} \langle \tau k_1, \bar{\tau} k_2 | V(r_1 - r_2) | \bar{\tau} k_4, \tau k_3 \rangle,$$

(A52)

$$V_{\text{int}}^{(7)} = \frac{1}{2N} \langle \tau k_1, \bar{\tau} k_2 | V(r_1 - r_2) | \tau k_4, \bar{\tau} k_3 \rangle,$$

(A53)

$$V_{\text{int}}^{(8)} = \frac{1}{2N} \langle \tau k_1, \bar{\tau} k_2 | V(r_1 - r_2) | \bar{\tau} k_4, \bar{\tau} k_3 \rangle.$$

(A54)

$\bar{\tau}$ ($\bar{s}$) represents the opposite valley (spin) of $\tau$ ($s$). $k$ ($\bar{k}$) indicates the relative wave vector with respect to the minimum of $\tau$ ($\bar{\tau}$) valley. As for $T_1$, we take $s_2 = \bar{s}_1$. Due to the Pauli exclusion principle, electrons with the opposite spin is apt to be spatially closer than those with the same spin. Therefore, the contribution of the term
\[ s_2 = s_1 \text{ is omitted. The momentum conservation is employed. In } T_1 \text{ term, we take } k_4 = k_1 \text{ and } k_3 = k_2. \text{ As for } T_6, \text{ we are focused on the spin exchange and neglect the momentum scattering in the process. Therefore, we take } k_3 = k_1 \text{ and } k_4 = k_2. \text{ It is convenient to define } U = \chi^{(1)} \text{ and } U' = \chi^{(6)}. \text{ The intravally and intervalley } e-e \text{ interaction are then written as}
\]
\[
\dot{V}_{\text{intra}} = \frac{1}{N} \sum_{\tau s} \sum_{k_1 k_2} U \left( a_{k_1 s}^\dagger a_{k_2 \bar{s}} a_{k_2 s}^\dagger a_{k_1 \bar{s}} \right),
\]
\[ (A55) \]
\[
\dot{V}_{\text{inter}} = \frac{1}{N} \sum_{\tau \tau' s_1 s_2} \sum_{k_1 k_2} U' \left( a_{k_1 s_1}^\dagger a_{k_2 s_2}^\dagger a_{k_2 s_1} a_{k_1 s_2} \right),
\]
\[ (A56) \]

Therefore, the total Hamiltonian is obtained
\[
\dot{H} = \dot{H}_0 + \dot{V}_{\text{intra}} + \dot{V}_{\text{inter}},
\]
\[ (A57) \]

which includes the intravally and intervalley interaction. In the following, \( \dot{H} \) is solved at the mean field level.

**Appendix B: mean field approximation**

As for \( \dot{V}_{\text{intra}} \), we take the mean field approximation (MFA) directly
\[
\dot{V}_{\text{intra}}^{\text{MF}} = \frac{1}{N} \sum_{\tau s} \sum_{k_1 k_2} U \left( \langle a_{k_1 s}^\dagger a_{k_2 s}^\dagger \rangle a_{k_2 \bar{s}} a_{k_1 \bar{s}}^\dagger \right.
\]
\[ + \left. a_{k_1 s}^\dagger a_{k_2 s} a_{k_2 \bar{s}}^\dagger a_{k_1 \bar{s}} \rangle - \langle a_{k_1 s}^\dagger a_{k_2 s}^\dagger \rangle \langle a_{k_2 \bar{s}} a_{k_1 \bar{s}} \rangle \right).
\]
\[ (B1) \]

In the mean field approximation, we neglect the second order quantum fluctuations. The third term in above equation is omitted, because it is a constant, which can not effect the following qualitative discussion of the result. MFA of \( \dot{V}_{\text{intra}} \) reads
\[
\dot{V}_{\text{intra}}^{\text{MF}} \approx \frac{1}{N} \sum_{\tau s} \sum_{k_1 k_2} U \left( \langle n_{k_2 \bar{s}} \rangle n_{k_1 s}^\dagger + n_{k_1 s}^\dagger \langle n_{k_2 \bar{s}} \rangle \right),
\]
\[
\approx \frac{2}{N} \sum_{\tau s} \sum_{k_1 k_2} U \langle n_{k_2 \bar{s}} \rangle n_{k_1 s},
\]
\[
\approx \sum_{\tau k s} U_{\tau s} a_{k_s}^\dagger a_{k_s}^\dagger,
\]
\[ (B2) \]

where
\[
U_{\tau s} = \frac{2}{N} \sum_k U \langle n_{k_s}^\dagger \rangle.
\]
\[ (B3) \]

\( n_{k_s}^\dagger = a_{k_s}^\dagger a_{k_s}^\dagger \) is the particle number operator. Here, we merely consider the zero temperature case. So, \( \langle \cdots \rangle \) is the ground state average. As for \( \dot{V}_{\text{inter}} \), we rewrite it in terms of the spin operators in order to extract the intervalley spin exchange interaction
\[
\dot{V}_{\text{inter}} = \frac{1}{N} \sum_{k_1 k_2} U' \left( n_{k_1}^\dagger n_{k_2}^\dagger + 4 S_{\tau k_1} \cdot S_{\tau k_2} \right).
\]
\[ (B4) \]

The spin coupling term reads
\[
S_{\tau k_1} \cdot S_{\tau k_2} = S_{\tau k_1}^z S_{\tau k_2}^z + \frac{1}{2} \left( S_{\tau k_1}^+ S_{\tau k_2}^- + S_{\tau k_1}^- S_{\tau k_2}^+ \right),
\]
\[ (B5) \]

where \( S_{\tau k} = \frac{1}{2} (n_{k_1} - n_{k_2}) \), \( S_{\tau k}^+ = a_{k_1}^\dagger a_{k_2}^\dagger \) and \( S_{\tau k}^- = a_{k_2}^\dagger a_{k_1}^\dagger \). It is obvious that the intervalley spin coupling is extracted. We chose the direction of \( S \) as the z-axis and apply MFA to Eq. (B4) obtaining
\[
\dot{V}_{\text{inter}}^{\text{MF}} = \frac{1}{N} \sum_{k_1 k_2} U' \left( \langle n_{k_1}^\dagger \rangle n_{k_2}^\dagger + \langle n_{k_2}^\dagger \rangle n_{k_1}^\dagger \right)
\]
\[
+ 4 \langle S_{\tau k_1}^z \rangle S_{\tau k_2}^z + 4 S_{\tau k_1}^z \langle S_{\tau k_2}^z \rangle - 4 \langle S_{\tau k_1}^z \rangle \langle S_{\tau k_2}^z \rangle \right)
\]
\[
\approx \sum_{\tau k} \left( U_{\tau}^s n_{k}^\dagger + M_{\tau} S_{\tau k}^z \right)
\]
\[
\approx - \sum_{\tau k} \left( U_{\tau}^s + \frac{1}{2} sM_{\tau} \right) n_{k}^\dagger
\]
\[
\approx - \sum_{\tau k s} X_{\tau s} a_{k_s}^\dagger a_{k_s}^\dagger
\]
\[ (B6) \]

where
\[
U_{\tau}^s = \frac{1}{N} \sum_k U' \langle n_{k_s}^\dagger \rangle,
\]
\[ (B7) \]

\[
M_{\tau} = \frac{4}{N} \sum_k U' \langle S_{\tau k}^z \rangle,
\]
\[ (B8) \]

\[
X_{\tau s} = U_{\tau}^s + \frac{1}{2} sM_{\tau}.
\]
\[ (B9) \]

Therefore, MFA of the interaction operator \( \dot{V} \) is
\[
\dot{V}_{\text{MF}} = \dot{V}_{\text{intra}}^{\text{MF}} + \dot{V}_{\text{inter}}^{\text{MF}}
\]
\[
= \sum_{k s} \left( F_{\tau s} a_{k_s}^\dagger a_{k_s} + F_{\tau s} a_{k_s}^\dagger a_{k_s}^\dagger \right),
\]
\[ (B10) \]

where the effective mean field is
\[
F_{\tau s} = U_{\tau}^s - X_{\tau s},
\]
\[ (B11) \]

At the mean field level, the total hamiltonian reads
\[
H^{\text{MF}} = \sum_{k s} \left( E_{c\tau s} (k) a_{k_s}^\dagger a_{k_s} + E_{c\tau s} (\bar{k}) a_{k_s}^\dagger a_{k_s}^\dagger \right),
\]
\[ (B12) \]

where the energy spectrum is
\[
E_{c\tau s} (k) = E_{c\tau s} (k) + F_{\tau s} - \mu.
\]
\[ (B13) \]

It is obvious that the effective mean field \( F_{\tau s} \) is obtained upon the calculation of \( \langle n_{k_s}^\dagger \rangle \). It is convenient to define
\[
\bar{n}_{s}^\dagger = \frac{1}{N} \sum_k \langle n_{k_s}^\dagger \rangle,
\]
\[ (B14) \]

which indicates the ratio of the occupation at \( \tau \) valley. Eq. (A32), Eq. (B3), Eq. (B7), Eq. (B8), Eq. (B9), Eq. (B11), Eq. (B13) and Eq. (B14) constitute a set of mean field self-consistent equations.
If \( A \) is the zone (BZ) of ML-MoS, it can be rewritten equivalently as the maximum of the angle. When \( 0 \leq k \) take the value of \( S \), we are able to solve the summation of the discrete values in Eq. (B14), we obtain

\[
\tilde{n}_s = \frac{3\sqrt{3}a^2 (k_F^{s})^2}{8\pi}.
\]  

(6)

When \( k_c < k_F^{s} \leq k_m \), \( S^{\tau s}_{\text{occ}} \) is divided into two parts: three triangle areas, three sectorial areas (see Fig. 6(b), regions divided by dash lines).

\[
S^{\tau s}_{\text{occ}} = S^{\tau s}_{\text{tri}} + S^{\tau s}_{\text{sec}}.
\]  

(7)

As for \( S^{\tau s}_{\text{tri}} \), we have

\[
S^{\tau s}_{\text{tri}} = 3k_c \sqrt{(k_F^{s})^2 - k_c^2}.
\]  

(8)

The sectorial area is

\[
S^{\tau s}_{\text{sec}} = \frac{3}{2} (k_F^{s})^2 \theta,
\]  

(9)

where \( \theta \) is the angle of the sector. Therefore,

\[
S^{\tau s}_{\text{occ}} = 3k_c \sqrt{(k_F^{s})^2 - k_c^2} + \frac{3}{2} (k_F^{s})^2 \theta.
\]  

(10)

Substituting into Eq. (C4), we have

\[
\tilde{n}_s = \frac{3a}{4\pi} \sqrt{(k_F^{s})^2 - k_c^2} + \frac{3\sqrt{3}}{12} \left( \frac{k_F^{s}}{k_c} \right)^2 \theta.
\]  

(11)

When \( k_F^{s} > k_m \), \( S^{\tau s}_{\text{occ}} \) is half of \( S_{\text{BZ}} \). Therefore, \( \tilde{n}_s = 1/2 \). Conclusively,

\[
\tilde{n}_s = \begin{cases} 
\frac{3a}{4\pi} \sqrt{(k_F^{s})^2 - k_c^2} + \frac{3\sqrt{3}}{12} \left( \frac{k_F^{s}}{k_c} \right)^2 \theta & 0 \leq k_F^{s} \leq k_c \\
\frac{1}{2} & k_c < k_F^{s} \leq k_m \\
k_m < k_F^{s} & k_m < k_F^{s}
\end{cases}
\]  

(12)

where

\[
\theta = 2\arcsin \left( \frac{\pi}{3ak_F^{s}} - \frac{1}{2k_F^{s}} \sqrt{(k_F^{s})^2 - k_c^2} \right).
\]  

(13)

As for the electron which fills the conduction band, its contribution to the free energy is defined by the integration

\[
E^{\tau s}_{\text{free}} = \int_{S^{\tau s}_{\text{occ}}} dk \left( \tilde{\alpha}_{rs} + \sqrt{(atk)^2 + \beta^2} \right).
\]  

(14)

Hence, the total free energy reads

\[
E_{\text{free}} = \sum_{\tau s} E^{\tau s}_{\text{free}}.
\]  

(15)

In Eq. (C14), we neglect the wave vector density. When \( 0 \leq k_F^{s} \leq k_c \), the region occupied by the electrons in BZ is a circular region. \( E^{\tau s}_{\text{free}} \) is calculated directly.
\[ E_{\text{free}}^s = - \int_{E_{\text{tau}}(k^2)}^{E_{\text{tau}}(k^2)} \pi k^2 dE \]
\[ = - \int_{E_{\text{tau}}(k^2)}^{E_{\text{tau}}(k^2)} \pi \left( \frac{E - A_{\tau s}}{2} \right)^2 dE \]
\[ = - \frac{\pi}{3(at)^2} \left( A_{\tau s}^3 - 2B_{\tau s}^3 - 2\tau s A_{\tau s} \right). \]  

In above derivation, we use \( E(k^2) = 0 \) and \( E(0) = A_{\tau s} + B_{\tau s} \). As for the case \( k_c < k_F \leq k_m \), \( E_{\text{free}}^s \) is composed of three parts, which are corresponding to the integration over the region \( s_1, s_2 \) and \( s_3 \) as shown in Eq. 6(b),

\[ E_{\text{free}}^s = E_{1}^s + E_{2}^s + E_{3}^s. \]  

We calculate the integration individually.

(1) The integration in region \( s_1 \) is

\[ E_{1}^s = - \int_{E_{\text{tau}}(k_c)}^{E_{\text{tau}}(k_c)} \pi k^2 dE - \int_{E_{\text{tau}}(k_c)}^{E_{\text{tau}}(k_c)} \pi k^2 dE \]
\[ = - \frac{\pi}{3(at)^2} \left( A_{\tau s}^3 - 2B_{\tau s}^3 - 2\tau s A_{\tau s} \right). \]  

(2) The integration in region \( s_2 \) is

\[ E_{2}^s = \frac{\theta}{2(at)^2} \left( A_{\tau s}^3 - 2B_{\tau s}^3 - 2\tau s A_{\tau s} \right) \]  

(3) The integration in region \( s_3 \) is complicated. We have

\[ E_{3}^s = 6 \int_{\tau s} A_{\tau s} + \sqrt{(akc)^2 + B_{\tau s}^2} dk_x dk_y \]
\[ = 6 \int_{\phi_m}^{\phi_m} d\phi \int_{k_c} k \left( A_{\tau s} + \sqrt{(akc)^2 + B_{\tau s}^2} \right) dk \]
\[ = 6 \int_{\phi_m}^{\phi_m} d\phi \left[ \frac{1}{2} A_{\tau s} k_c^2 \left( \frac{\sin^2(\phi)}{\cos(\phi)} - 1 \right) \right] \]
\[ + \frac{1}{3(at)^2} \left( \frac{(akc)^2 + B_{\tau s}^2}{\cos(\phi)} \right)^{3/2} \]
\[ - \frac{1}{3(at)^2} \left( (akc)^2 + B_{\tau s}^2 \right)^{3/2} \]
\[ = 3A_{\tau s} k_c^2 (\cos(\phi) - \phi_m) - 2(\tau s) c_{\tau s} \]
\[ + 6D_{\tau s}, \quad \text{(C21)} \]

where \( \phi_m = \arccos(\sqrt{\frac{c_{\tau s}}{k_c^2}}) \). \( D_{\tau s} \) denotes an integration

\[ D_{\tau s} = \frac{1}{3(at)^2} \int_{\phi_m}^{\phi_m} d\phi \left[ \left( \frac{\cos(\phi)}{\cos(\phi)} \right)^2 + B_{\tau s}^2 \right]^{3/2}. \]  

It is hard for \( D_{\tau s} \) to obtain an analytical formula. Therefore, \( D_{\tau s} \) is calculated numerically. When \( k_c < k_F \leq k_m \),

\[ E_{\text{free}}^s = \frac{\pi}{3(at)^2} \left( 3A_{\tau s}^3 - C_{\tau s}^3 - 2B_{\tau s}^3 \right) + \pi k_c^2 E_{\text{crs}}(k_c) \]
\[ + \frac{\theta}{2(at)^2} \left( A_{\tau s}^3 - 2B_{\tau s}^3 - 3\tau s A_{\tau s} \right) - \frac{\theta}{2\tau} E_{\tau s}^s \]
\[ + 3A_{\tau s} k_c^2 (\cos(\phi) - \phi_m) - 2(\tau s) c_{\tau s} \]
\[ + 6D_{\tau s}. \]  

As for \( 0 \leq k_F^\ast \leq k_c \), the total free energy is obtained by substituting Eq. (C16) into Eq. (C15). For \( k_c < k_F^\ast \leq k_m \), the total free energy is calculated by substituting Eq. (C17) into Eq. (C15).
In our model Hamiltonian, we merely consider two conduction and valence bands. Since the band edges of conduction and valence bands are renormalized by electron Coulomb interaction and shifted from those in non-interacting case, the chemical potentials studied in this work for various figures are finally not lying deeply into conduction band after the renormalization. In this sense, the effect of more bands with higher energies cannot play important role in this study. Moreover, we believe that the strong electron-electron interaction at low electron density plays a determining role in the ferromagnetic ground state. The high chemical potential leads to more other conduction bands occupied and high electron density [30], which can’t change the phase transition discussed in this paper.

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