Spin-valley half-metal as a prospective material for spin-valley-tronics

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Half-metallicity (full spin polarization of the Fermi surface) usually occurs in strongly correlated electron systems. We demonstrate that doping a spin-density wave insulator in the weak-coupling regime may also stabilize half-metallic states. The undoped spin-density wave is formed by four nested bands [i.e., each band is characterized by charge (electron/hole) and spin (up/down) labels]. Of these four bands only two accumulate the doped carriers, forming a half-metallic two-valley Fermi surface. Depending on parameters, the spin polarizations of the electron-like and hole-like valleys may be (i) parallel or (ii) antiparallel. The Fermi surface of (i) is fully spin-polarized (similar to usual half-metals). Case (ii), referred to as “a spin-valley half-metal”, corresponds to complete polarization with respect to the spin-valley operator. The properties of these states are discussed.

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Introduction.— Half-metallicity [1–3] is a useful property for spintronics applications. Unlike usual metals, which has both spin projections (spin-up and spin-down) on the Fermi surface, half-metallicity implies that the electrons with only one spin projection, for example, spin-up, reach the Fermi level. Spin-down states are pushed away from the Fermi energy, making the ground state non-invariant under spin-rotation transformations. A highly desirable consequence [3, 4] of half-metallicity is the perfect spin-polarization of electric current. Experimental studies confirmed that a variety of real materials are half-metals. For example, NiMnSb [3]; (La0.7Sr0.3)MnO3 [4]; CrO2 [7], Co2MnSi [8], as well as many others.

From the theory standpoint, the half-metallicity of these compounds relies on sizable electron-electron interactions, which are associated with transition metal atoms. However, in recent years, the search for ‘metal-free half-metals’ began [9, 10]. Such systems could be useful for bio-compatible applications, and, in general, are consistent with current interest in carbon-based and organic-based mesoscopic systems [11–14]. It is difficult to expect a strong electron-electron interaction for systems composed entirely of s- and p-elements. Thus, different mechanisms for half-metallicity must be looked for.

In this paper, we discuss a novel possibility to generate a half-metallicity. In brief, we demonstrate that doping a spin-density wave (SDW) insulator may stabilize a type of half-metallic state. Let the undoped system [see Fig. 1(a)] have two nested Fermi surface sheets, which we will also refer to as valleys. Let one sheet, or valley, correspond to electron states, and another to hole states. Both valleys are spin-degenerate. The SDW instability opens a gap generating an insulating ground state. We show that, when doping is introduced, each valley becomes half-metallic. If the spin polarizations of both sheets are parallel to each other, Fig. 1(c), a half-metallic state, called below charge-density wave (CDW) half-metal, emerges. For antiparallel polarizations, Fig. 1(d), a different half-metallic state, spin-valley half-metal, appears. The properties of these two states are discussed below.

Model.— Our model has two bands, or valleys: an
electronic band $a$ and a hole band $b$, Fig. [1(a)], with the following single-particle dispersion ($\hbar = 1$)

$$
\varepsilon^a(k) = \frac{k^2}{2m_a} + \varepsilon^a_{\min} - \mu, \quad \varepsilon^a_{\min} < \varepsilon^a < \varepsilon^a_{\max},
$$

(1)

$$
\varepsilon^b(k + Q_0) = -\frac{k^2}{2m_b} + \varepsilon^b_{\max} - \mu, \quad \varepsilon^b_{\min} < \varepsilon^b < \varepsilon^b_{\max}.
$$

(2)

Here band $a$ is centered at $k = 0$, and band $b$ at some finite momentum $Q_0$. Below, to simplify calculations, we will assume perfect electron-hole symmetry: $m_a = m_b = m$, and $\varepsilon^a_{\max} = -\varepsilon^b_{\min} = \varepsilon_F$. Consequently, $\varepsilon^a(k) = -\varepsilon^b(k + Q_0) = \varepsilon_k$. Zero doping corresponds to $\mu = 0$. Undoped Fermi surface sheets for the $a$ and $b$ bands are characterized by a single Fermi momentum $k_F = \sqrt{2m \varepsilon_F}$, and density of states (per spin projection) $N_F = mk_F/(2\pi^2)$ at the Fermi energy. This provides a perfect nesting: a translation of the electron Fermi surface by the vector $Q_0$ completely superposes the sheets. The total Hamiltonian is equal to

$$
\hat{H} = \hat{H}_e + \hat{H}_{\text{int}},
$$

(3)

where $\hat{H}_e$ is the single-electron term, described by the dispersions (12), while $\hat{H}_{\text{int}}$ corresponds to the interaction between quasiparticles.

To treat the SDW instability, it is sufficient to keep in $\hat{H}_{\text{int}}$ only the interaction between electrons in the $a$ and $b$ bands. We also assume that the interaction is a short-range one. We will focus mostly on the following interaction term

$$
\hat{H}_{\text{int}} = g \int d^2x \sum_{\sigma, \sigma'} \psi^\dagger_{a\sigma}(x) \psi_{a\sigma}(x) \psi^\dagger_{b\sigma'}(x) \psi_{b\sigma'}(x).
$$

(4)

The effects of the neglected term will be discussed later. In Eq. (4) symbol $\psi_{a\sigma}$ denotes the usual fermionic field operator for band $a$ and spin $\sigma$. The coupling constant $g$ is positive, which corresponds to repulsion, and weak $gN_F \ll 1$.

**Spin-valley half-metal.** — When the Fermi surface sheets of the holes and the electrons perfectly match each other, model describes spontaneous formation of the SDW or CDW orders. We start with the SDW. The SDW ground state is believed to be unique (up to rotation of the spin polarization axis), and well-described by a BCS-like mean-field theory. The electron operators can be grouped into two sectors, labeled by the index $\sigma = \pm 1$: sector $\sigma$ consists of $\psi_{a\sigma}$ and $\psi_{b\bar{\sigma}}$ (here $\bar{\sigma}$ means $-\sigma$). In the mean-field approach the sectors are decoupled and the SDW order parameter can be written as

$$
\Delta_\sigma = \frac{g}{V} \sum_k \langle \psi^\dagger_{k\sigma} \psi_{k\bar{\sigma}} \rangle,
$$

(5)

where $V$ is the system volume, and $\langle \ldots \rangle$ denotes the diagonal matrix element for the ground state. At zero doping, the sectors are degenerate: $\Delta_\uparrow = \Delta_\downarrow = \Delta_0$, where $\Delta_0$ is the order parameter at perfect nesting. This equality implies that the SDW polarization in real space is along the $x$-axis:

$$
\langle S^x(r) \rangle = \frac{\Delta_\uparrow + \Delta_\downarrow}{g} \cos(Q_0r) = \frac{2\Delta_0}{g} \cos(Q_0r),
$$

(6)

$$
\langle S^y(r) \rangle = \frac{\Delta_\uparrow - \Delta_\downarrow}{2g} \sin(Q_0r) = 0.
$$

(7)

For $\Delta_0$ one can write $\Delta_0 \approx \varepsilon_F \exp(-1/gN_F)$.

Doping destroys the perfect Fermi surface nesting and the number of low-energy states competing to become the true ground state sharply grows. Both incommensurate and inhomogeneous phases [17–25] were considered for Hamiltonian [3] and its numerous variations. Here we argue that the half-metallic state is yet another viable contender in the case of imperfect nesting.

The grand potential of our system $\Omega$ at zero temperature and finite doping $x$ is a sum of two partial grand potentials $\Omega = \sum_\sigma \Omega_\sigma$, where

$$
\frac{\Omega_\sigma}{V} = \frac{\Delta_\sigma^2}{g} - \frac{1}{V} \sum_k \left( \mu + \varepsilon^2_k + \Delta_\sigma^2 \right) \left( \mu - \varepsilon^2_k + \Delta_\sigma^2 \right) + \left( \mu - \varepsilon^2_k + \Delta_\sigma^2 \right) \theta \left( \mu - \varepsilon^2_k + \Delta_\sigma^2 \right),
$$

(8)

and $\theta(z)$ is the Heaviside step-function. To describe doping it is convenient to introduce $x_\sigma = -\partial \Omega_\sigma / \partial \mu$. The physical meaning of partial doping $x_\sigma$ is the amount of doped charge accumulated in sector $\sigma$. The value of $\Delta_\sigma$ corresponds to the minimum of $\Omega_\sigma$. Thus, one has to solve the system of equations

$$
\frac{\partial \Omega_\sigma}{\partial \Delta_\sigma} = 0, \quad x_\uparrow + x_\downarrow = x,
$$

(9)

to determine $\mu$ and $\Delta_\sigma$. Equations (8, 9) are valid provided that (a) the state remains homogeneous, and (b) the SDW order parameter remains commensurate even at finite doping.

Since the two sectors $\sigma$ are decoupled, one can calculate $\Delta_\sigma$ and $\mu$ as functions of $x_\sigma$. The result is [18, 25, 26]

$$
\Delta_\sigma = \Delta_0 \sqrt{1 - \frac{x_\sigma}{N_F \Delta_0}}, \quad \mu = \Delta_0 - \frac{x_\sigma}{2N_F}.
$$

(10)

Thus, the order parameter decreases at finite doping, and the homogeneous commensurate state becomes completely unstable for $x_\sigma > x_c = N_F \Delta_0$.

It is often implicitly assumed (see, e.g., Refs. [17, 25, 26]) that the charge carriers are spread evenly between both sectors of $\sigma$, and the degeneracy of $\Delta_\sigma$ persists even for finite $x$. Yet, it is easy to show that the spontaneous lifting of this degeneracy optimizes the energy. To prove this, consider the system’s free energy $F = \sum_\sigma F_\sigma$, where the partial free energy $F_\sigma = \Omega_\sigma + \mu x_\sigma$ can be calculated as $F_\sigma(x_\sigma) = F_\sigma(0) + \int_0^{x_\sigma} d\bar{x} \mu(\bar{x})$. One thus obtains

$$
F = \sum_\sigma \frac{F_\sigma}{V} = -N_F \Delta_0^2 + \Delta_0 x - \frac{x^2}{4N_F} + \frac{1}{4N_F},
$$

(11)
where we took into account that $F_\sigma(0) = -N_F\Delta_0^2/2$. Note that only the third term in Eq. (11) depends on the distribution of the dopants among the two sectors. It is easy to check that, if $x_\sigma = x$ and $x_\bar{\sigma} = 0$, the third term, together with $F$, is the smallest. In other words, for fixed $x$, the most stable spatially-homogeneous state of the model corresponds to the case when all the doped charge accumulates in a given sector. The other sector is completely free of the extra charge carriers. Therefore

$$\frac{\mu}{\sigma} = \Delta_0 - \frac{x}{2N_F}, \quad \Delta_\sigma(x) = \Delta_0 \sqrt{1 - \frac{x}{N_F\Delta_0}}.$$  

These relations are valid for not too strong doping $x > N_F\Delta_0$. An important feature of Eq. (12) is that the second derivative $\delta^2 F/\delta x^2$ is negative. This means that the doped system is unstable with respect to electronic phase separation [13, 22, 23, 24, 27]. However, the Coulomb interaction can suppress the phase separation [28, 29]. Thus, it is reasonable to study the properties of the homogeneous state.

It follows from Eqs. (13, 14) that $\Delta_\sigma(x) < \mu(x) < \Delta_0(x) = \Delta_0$, when $x > 0$. This means that in the sector $\bar{\sigma}$, the order parameter remains equal to $\Delta_0$ and, since the chemical potential is lower than $\Delta_\sigma$, no charge enters this sector, see Fig. (1d). In the sector $\sigma$, two Fermi surface sheets emerge. They are fixed by the equation $\varepsilon_k^2 = [\mu(x)]^2 - [\Delta_\sigma(x)]^2$, which is equivalent to

$$\varepsilon_k = \pm \frac{x}{2N_F}.$$  

As the doped charges are distributed unevenly between the sectors, the doped state acquires non-trivial macroscopic quantum numbers. To characterize the macroscopic state, it is useful to specify spin ($\hat{S}$) and spin-valley ($\hat{S}_v$) operators

$$\hat{S} = \sum_{\alpha\sigma} \sigma \hat{N}_{\alpha\sigma}, \quad \hat{S}_v = \sum_{\alpha\sigma} \sigma v_\alpha \hat{N}_{\alpha\sigma},$$  

where $\alpha = a, b$, and the valley index $v_\alpha$ is defined as follows: $v_a = 1$ and $v_b = -1$. The operator $\hat{N}_{\alpha\sigma}$ counts the number of electrons with spin $\sigma$ in valley $\alpha$

$$\hat{N}_{\alpha\sigma} = \sum_k \psi_{k\alpha\sigma}^\dagger \psi_{k\alpha\sigma}.$$  

The Hamiltonian [3] commutes with both $\hat{S}$ and $\hat{S}_v$. The field operators satisfy obvious commutation rules

$$[\hat{S}, \psi_{\alpha\sigma}] = \sigma \psi_{\alpha\sigma}, \quad [\hat{S}_v, \psi_{\alpha\sigma}] = \sigma v_\alpha \psi_{\alpha\sigma}.$$  

In other words, in addition to the spin quantum number $\sigma$, a field $\psi_{\alpha\sigma}$ can be characterized by the spin-valley projection $\sigma v_\alpha$.

It is easy to check that in the sector $\sigma$ both $\psi_{a\sigma}$ and $\psi_{b\bar{\sigma}}$ carry the same spin-valley quantum equal to $+\sigma$. In the sector $\bar{\sigma}$, the field operators correspond to a $-\sigma$ quantum of $\hat{S}_v$. That is, the Fermi surface of the doped system is characterized by the single projection of the spin-valley operator. The Fermi surface sheets with the opposite projection of $\hat{S}_v$ are absent, since the sector $\sigma$ is gapped. Thus, the doped system can be referred to as a spin-valley half-metal: like a classical half-metal, our system exhibits complete polarization of the Fermi surface; however, in contrast to the usual half-metal, the polarization is not the spin polarization, but rather, the spin-valley one. Therefore, the electrical current through the spin-valley half-metal is completely spin-valley polarized.

Since the sector $\bar{\sigma}$ is free from doped electrons, the average values of $\hat{N}_{a\bar{\sigma}}$ and $\hat{N}_{b\bar{\sigma}}$ remain unaffected by the doping, while $\langle N_{a\sigma} \rangle$ and $\langle N_{b\sigma} \rangle$ change. Taking the average occupation numbers $N_{a\sigma} = \langle \hat{N}_{a\sigma} \rangle$ in the undoped state to be zero, we can write $N_{a\bar{\sigma}} = N_{b\sigma} = 0$, and $N_{a\sigma} + N_{b\bar{\sigma}} = x V$. Consequently, $S_v = \langle \hat{S}_v \rangle$ is proportional to $x$. Namely, $S_v = x \sigma V$. In a system with the perfect electron-hole symmetry we have $N_{a\sigma} = N_{b\bar{\sigma}} = V\Delta_0/2$, which corresponds to $\hat{S}_v = 0$, for any $x$. When the symmetry is absent: $|S| \propto x$. However, the net spin polarization of the spin-valley half-metal satisfies the inequality $|S| < |S_v|$.

Doping also affects the SDW order inherited from the undoped state. Intuitively, since the charge enters only one of the two sectors, the symmetry between sectors $\sigma$ disappears for $x > 0$. [Eqs. (14) prove this.] Thus, simple SDW is replaced by a more complicated order parameter: analyzing Eqs. (9, 10) one can prove that, at finite doping, a circularly-polarized spin component emerges

$$\{\delta S^x(r), \delta S^y(r)\} \propto \langle \Delta_1 - \Delta_\bar{\sigma} \rangle \{\cos(Q_0 r), \sin(Q_0 r)\}. \quad (19)$$  

The amplitude of this component increases when $x$ grows.

From spin-valley half-metal to CDW half-metal. — In addition to the expected invariance with respect to simultaneous rotations of all fermion spins, our model Hamiltonian allows for a broader class of symmetries: it remains unchanged, even if the electron and hole spins are transformed by two different rotation operators. This observation can be trivially proven in the absence of interaction ($g = 0$). In the case of a generic interaction, this symmetry does not apply. However, if the interaction is short-range, as in Eq. (14), the invariance of the Hamiltonian under such transformations remains. Indeed, the integrand in Eq. (14) is $\propto \rho_e \rho_h$, where $\rho_e$ and $\rho_h$ are the density operators for electrons and holes, which both are invariant under separate rotations of the electron and hole spins. Therefore, the substitution

$$\psi_{\bar{\alpha}\bar{\sigma}} \to \psi_{b\bar{\sigma}}, \quad \psi_{a\bar{\sigma}} \to \psi_{b\bar{\sigma}}$$  

corresponds to a symmetry of the model. Consequently, Eq. (20) either preserves the ground state, or transforms
one ground state into another one. Since the order parameter, Eq. (19), changes under the transformation (20), we must conclude that a new ground state is generated by such a substitution. If we start with the spin-valley half-metal ground state, what kind of new state the transformation (20) brings us?

Consider the SDW polarization, Eq. (16), at zero doping. Under the transformation (20) the SDW is replaced by charge-density wave with finite average value of the density operator \( \hat{\rho}_{Q_0} \):

\[
\langle \hat{S}_{Q_0} \rangle = \sum_{\sigma} \langle \hat{\psi}_{\alpha \sigma}^\dagger \hat{\psi}_{\beta \sigma} \rangle \to \sum_{\sigma} \langle \hat{\psi}_{\beta \sigma} \hat{\psi}_{\alpha \sigma} \rangle = \langle \hat{\rho}_{Q_0} \rangle.
\tag{21}
\]

Calculations identical (up to relabeling) to the case of the SDW order demonstrate that for \( x > 0 \) the charge carriers accumulate in a single mean-field sector. However, the sector composition is changed by the transformation (20): sector \( \sigma \) consists of \( \hat{\psi}_{\alpha \sigma} \) and \( \hat{\psi}_{\beta \sigma} \). Unlike the case of spin-valley half-metals, now both electronic fields within a single sector have the same spin projection. Therefore, if the doped charge enters sector \( \sigma \), both Fermi surface sheets have identical spin polarizations equal to \( \sigma \), see Fig. 1(c). This perfect polarization of the Fermi surface is a hallmark feature of half-metals. Thus, the spin-valley half-metal is related to the CDW half-metal by the substitution (20), and both states are degenerate within our model. This connection becomes apparent if we notice that (20) switches the operators \( \hat{S}_{Q_0} \) and \( \hat{S}_{\tau} \). Consequently, in the CDW half-metal \( S = x \sigma V \) and \( |S_\tau| < |S| \). When \( x > 0 \), in addition to the CDW order parameter, the SDW order parameter \( \langle \hat{S}_{Q_0} \rangle \) is generated. It grows monotonously as \( x \) grows. This is a direct analog of Eq. (19).

Note, however, that the degeneracy between the SDW and CDW ground states is an artifact of the short-range separation. In addition, the incommensurate SDW states are absent in more realistic models. The effects of more generic interaction operators are discussed below.

Discussion. — The interaction Eq. (4) is not the most general form of electron-electron coupling. In particular, the “exchange” term

\[
\hat{H}_{\text{ex}} = g_\perp \int d^3x \sum_{\sigma \sigma'} \langle \hat{\psi}_{\alpha \sigma} \hat{\psi}_{\beta \sigma'} \rangle \langle \hat{\psi}_{\beta \sigma'} \hat{\psi}_{\alpha \sigma} \rangle
\tag{22}
\]

should be accounted for. The coupling constant \( g_\perp > 0 \) describes a repulsive interaction at finite momentum \( Q_0 \). The “exchange” term (22) immediately lifts the degeneracy between the SDW and CDW, in favor of SDW. This means that, for finite doping, the spin-valley half-metal is more stable than the CDW half-metal. Also, other factors could favor the CDW half-metal, for example, the proximity to a lattice instability. An external magnetic field acts similarly, since the total spin of the CDW half-metal exceeds the spin of the spin-valley half-metal.

We assumed that the Coulomb interaction guarantees the homogeneity of the electron liquid. Thus, in the above discussion we neglected the possibility of phase separation. In addition, the incommensurate SDW states were not considered. While the detailed study of such states is an interesting goal for future research, we do not expect that this modification would affect significantly the stability of the half-metallic phases, at least at some doping region. Indeed, at the mean-field level the free energy in the presence of the incommensurate SDW equals

\[
F^\text{ic}(x) = \min_{x_\uparrow + x_\downarrow = x} \left[ F^\text{ic}_0(x_\uparrow) + F^\text{ic}_0(x_\downarrow) \right],
\tag{23}
\]

where \( F^\text{ic}_0(x_\uparrow) \) is the free energy of a sector with partial doping \( x_\uparrow \). As above, the free energy of the system is found by minimization under the condition \( x_\uparrow + x_\downarrow = x \). We calculated \( F^\text{ic}_0(x_\uparrow) \) numerically, as described in Ref. 26. Our analysis shows that \( \partial^2 F^\text{ic}_0(x_\uparrow)/\partial x^2 < 0 \) for \( x_\uparrow \) less than the threshold value \( x^* \approx 0.83 N_F \Delta_0 \). This is a rather general feature of a system with imperfect nesting [15, 22, 23, 26, 27]. Since the second derivative of \( F^\text{ic}_0 \) is negative, the sum \( F^\text{ic}_0(x_\uparrow) + F^\text{ic}_0(x_\downarrow) \) as a function of \( x_\uparrow \in [0, x] \) is concave. Consequently, the extremum of the latter sum at \( x_\uparrow = x/2 \) corresponds to a maximum, not a minimum (see Fig. 2). Thus, the total free energy is minimized as follows

\[
F^\text{ic}(x) = F^\text{ic}_0(x) + F^\text{ic}_0(0), \quad \text{at} \ x_\uparrow = x, \ x_\downarrow = 0.
\tag{24}
\]

Undoped sector \( \bar{\sigma} \) remains insulating. All doped charge goes to sector \( \sigma \), which becomes metallic, with a well-defined Fermi surface. Thus, we recover the spin-valley half-metal with an incommensurate SDW.

If \( x_\sigma > x^* \), then \( \partial^2 F^\text{ic}_0(x_\sigma)/\partial x^\sigma_\sigma > 0 \), and the total free energy \( F^\text{ic}_0(x_\sigma) + F^\text{ic}_0(x - x_\sigma) \) acquires a local minimum at \( x_\sigma = x_\uparrow = x/2 \) (see Fig. 2). When doping increases even further, this minimum becomes a global minimum for \( x \approx 1.8 N_F \Delta_0 \). Thus, the first order transition from incommensurate spin-valley half-metal to common incommensurate SDW phase occurs at this point.
We assume that both the electron and hole sheets in the Fermi surface have a spherical shape and are perfectly nested at zero doping. If these shapes are different, the range of doping where $\partial^2 F_0^{\text{nic}}(x)/\partial x^2 < 0$ diminishes [23]. When the geometry of the sheets differs significantly, one has $\partial^2 F_0^{\text{nic}}(x)/\partial x^2 > 0$ for all $x$, and the half-metal states become impossible.

To conclude, we demonstrated that doping a SDW state with perfectly-nested Fermi surface sheets stabilizes a half-metal-like ground state. Depending on microscopic parameters and the external magnetic field, such ground state could be either CDW half-metal with complete spin-polarization of the Fermi surface or spin-valley half-metal. The Fermi surface of the latter state is characterized by a perfect polarization in the spin-valley space. While the CDW half-metal supports purely spin-polarized currents, which is a natural consequence of the Fermi surface polarization, the spin-valley half-metal supports spin-valley-polarized currents.

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