Magnetochiral Polarization and High Order Transport in DNA type Chiral Materials

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The chirality–spin interaction is a fascinating topic for both physicists and chemists. For example, chiral molecules like DNA generate giant spin polarization in nanodevices characterized by large magnetoresistance. This phenomenon called chiral induced spin selectivity (CISS) in literature, paves pathways for unconventional spintronic devices and enantiomer separation. However, its physical mechanism is elusive and debated. In this work, we find that the CISS magnetoresistance is a high-order effect and originates from the current-driven charge accumulation at metal-chiral molecule interfaces. Reversing lead magnetization modulates the charge polarization and consequently changes the tunneling barrier across the molecule. The magnetoresistance increases with the barrier width and bias and can theoretically approach 100%. This mechanism can be validated by examining the insulation of chiral molecules, spin-orbit coupling in leads, or frequency-dependence of surface potential. Further, we propose that emerging twisted van der Waals quantum materials will be a versatile platform for CISS and similar spin selective phenomena.

1 Introduction

Chirality is a fundamental concept in chemistry, physics, and biology. Recently, chirality was reported to generate intriguing spin polarization, called chiral induced spin selectivity (CISS).
CISS opens exciting pathways to design unconventional spintronic devices using chiral molecules, perform chiral separation via magnetism, and explore the spin-selective biological process. However, the physical mechanism underlying CISS is elusive and debated.

In experiments, CISS is commonly probed by the two-terminal magnetoresistance (MR) where chiral molecules connect with a magnetic substrate and a nonmagnetic tip (see Figure 1a). The resistance changes as flipping the substrate magnetization ($M$), as illustrated in Figure 1c. Because the magnetoresistance is similar to a spin valve, chiral molecules were usually presumed to induce spin polarization. CISS MR increases with bias and molecule length and can be larger than 60%, although the Ni electrode exhibits only 20% spin polarization.

Furthermore, the CISS MR exhibits fundamentally distinct symmetry in the I-V curves (see Figures 1c and 1d) from another known chiral induced phenomenon called electric magnetochiral anisotropy. The magnetochiral anisotropy refers to the resistance that depends on the current ($I$) and magnetic field ($B$) for a chiral conductor, $R^\chi = R_0(1 + \alpha B^2 + \beta^\chi B \cdot I)$, where $\beta^\chi = -\beta^{-\chi}$ and $\chi$ stands for chirality. The $B^2$ term represents the ordinary MR while the $B \cdot I$ term represents the unidirectional resistance like a diode, for which we dub a magnetochiral diode (MCD). The MCD behavior was observed in many chiral materials (see Ref.15 for review and references therein), where the MR usually reaches several percentages and is much smaller than CISS MR. Corresponding I-V relation expands to the $V^2$ term while CISS requires a significant $V^3$ term. The magnetochiral anisotropy is a second-order effect and obeys the Onsager’s reciprocal relation. But CISS is an even higher-order effect. Onsager’s reciprocal theorem originates in
the microscopic reversibility of thermodynamic equilibrium and posed strict constraints on the macroscopic conductivity that is derived by perturbing the equilibrium state. The reciprocity requires that two-terminal resistance remains unchanged as reversing time \((B/M \rightarrow -B/-M, I \rightarrow -I)\), i.e., \(R(B/M, I) = R(-B/-M, -I)\) \(^{16,17}\). It holds for the MCD \(^{14,15}\) but not for CISS. In typical I-V curves, MCD is antisymmetric for reversing \(M\) while CISS looks nearly antisymmetric for the fixed \(M\) (see Figures 1c-1d) \(^{18–20}\).

The reciprocity violation indicates that CISS MR is a higher-order effect, which is driven by highly nonequilibrium phases. The flowing current may induce distinct nonequilibrium states for opposite magnetization and consequently generate different resistance. If so, what kind of nonequilibrium states appear? Recently, CISS was also proposed to exhibit MCD-like \(I-V\) relation to the second-order of \(V\) \(^{18,20}\), because a CISS device and an MCD share the same symmetry-breaking, both inversion-breaking and time-reversal symmetry-breaking. More specifically, however, what causes different transport behaviors between CISS and an MCD? Why does CISS exhibit higher-order effects than the MCD? Answers to these questions will be helpful to understand the nature of CISS. In experiments, an MCD involves conductors or doped semiconductors while CISS relates to insulating chiral molecules. For example, the resistance is in the order of magnitude \(10^9\) \(\Omega\) (current is \(\sim 1\) nA at bias 1 V) in CISS experiments \(^7\). We point out that the insulating nature of chiral molecules may be relevant for CISS MR, which was rarely appreciated in literature.

In this article, we propose that the CISS MR originates in the current-induced charge polarization at lead-molecule interfaces. The accumulated charge sensitively modulates the tunneling
Figure 1: **The chiral induce spin selectivity (CISS) device and symmetry of magnetoresistance.**

(a) Schematics of a two-terminal device to measure CISS. Switching substrate magnetization ($M$) leads to magnetoresistance. (b) The effective circuit diagram. A magnetochoiral diode ($D^\chi_M$) is in serial to an RC circuit and represents influences of chirality and magnetism. (c)-(d) Illustration of typical I-V curves for CISS and the electric magnetochoiral anisotropy. The curve for $M(-M)$ is shown by solid red (dashed blue) lines. The electric magnetochoiral anisotropy respects the Onsager’s reciprocal relation and is a second-order effect. In contrast, CISS violates the reciprocity and the I-V relation involves at least the third order of $V$. The magnetoresistance (MR) is usually estimated by $MR = \frac{I_{+M} - I_{-M}}{I_{+M} + I_{-M}}$ at given bias.
potential barrier through the insulating chiral molecule. We demonstrate that the MCD (see the diode in Figure 1b) leads to a correction to the charge accumulation. Because it flips in sign when reversing magnetization, the charge correction leads to magnetization-dependent nonequilibrium states and breaks the microscopic reversibility. Thus, MR refrains from the Onsager’s relation and turns into a higher-order effect. Since charge polarization is stabilized by the insulating layer, we predict that CISS MR will diminish and evolve into the electric magnetochiral effect when chiral molecules become metallic. Because it is sensitively determined by the tunneling barrier that is modified by the accumulated charge, MR is not constrained by the spin polarization rate in lead. The MR dependence on molecule length is well reproduced, too. Our theory shows that CISS is a highly nonlinear phenomenon, different from spin valves and the electric magnetochiral anisotropy. In addition, the magnetochiral charge correction also explains the work function experiment on a ferromagnetic film covered by chiral molecules. We further anticipate that the work function change is a dynamical property and depends on the measurement frequency.

2 Results

Because MR rather than spin polarization is the directly measured property in CISS experiments, we circumvent the vague spin polarization and focus on MR from general symmetry principles. Because of both inversion and time-reversal breaking, a CISS device leads to the MCD effect in the second-order conductivity, which is independent of model details such as spin-orbit coupling (SOC) or phonons. In CISS, we will treat the third-order response as a consequence of the second-order effect, by exploring the charge accumulation due to MCD. Subsequently, we
demonstrate that the charge accumulation modifies the tunneling barrier and consequently leads to a large MR.

**Current-induced charge accumulation** In experiments, two leads sandwich a layer of chiral molecules that are highly insulating. When a small current passes through the high-resistance layer, local charge polarization is not fully screened and accumulates at metal-molecule interfaces, presenting quantum capacitance\(^{23,24}\). Thus, we mimic the device with a parallel resistor-capacitor (RC) circuit. We note that a similar RC circuit was extensively adopted to study superconductor junctions\(^ {25}\). Because the device acts as an MCD in the second-order response, we introduce an MCD in serial to the RC circuit to simulate the experiment, as shown in Figure 1b. We will focus on the influence of MCD on capacitor charging. For example, after turning on a DC bias, the capacitor gets charged from zero to \(Q\) and the circuit develops into a steady state after a short time. The existence of MCD can modify \(Q\) via the current rectification.

Next, we demonstrate MCD-modified charge accumulation by simple circuit calculations, where we temporarily assume the total resistance as a constant. The I-V relation of MCD is treated as\(^ {20}\)

\[
I_D = (V_D + D_M^X V_D^2)/R_D, \tag{1}
\]

where \(V_D\) is the bias drop on MCD, \(R_D\) is the ordinary resistance, and \(D_M^X\) represents the magnetochiral current rectification, i.e., \(D_M^X = -D_M^{-X} = -D_M^\pm\). Suppose a total bias \(V_b\), the current and
Figure 2: **Charge accumulation** ($Q$) in a CISS device. (a) The RC circuit connects to opposite magnetochiral diodes, $D^X_M$ and $D^X_{-M}$. (b) By applying a DC bias, the time evolution of $Q$ shows $D^X_M$ induces more charge than $D^X_{-M}$ by $\Delta Q$. $\Delta Q$ is independent of the bias ($V_0$) direction. Dashed lines correspond to the case of $D^X_M = 0$. (c) By applying an AC bias $V = V_0 \sin (2\pi ft)$, $\Delta Q$ at peak positions remains the same as the DC case, also independent of the bias sign. Here, the low-frequency limit holds for $2\pi f \ll 1/RC$. (d)-(f) The $\Delta Q$ dependences on $V_0$, $D^X_M$, and $R$, which are the same for DC and AC simulations.
capacitor charge satisfy following conditions,

\[ I_D = \frac{dQ}{dt} + \frac{Q}{CR}, \quad (2) \]

\[ Q = C(V_b - V_D), \quad (3) \]

where \( R \) is the resistance, \( C \) the capacitance and \( Q \) the capacitor charge. For time-varying \( V_b \), we simulate the time-evolution of \( Q \) by numerically solving nonlinear Eqs.1-3.

We first conceive a simple DC voltage \( V_b(t) = \begin{cases} V_0 t/t_s, & 0 \leq t < t_0 \\ V_0, & t \geq t_0 \end{cases} \) where \( t_0 \) is the turning on time and \( V_0 \) is the steady bias. In the steady-state, \( Q \) is always larger for \( D_x^M \) than for \( D_{-M}^x \) by \( \Delta Q \), as shown in Figure 2b. We stress that \( \Delta Q \) is independent of the \( V_0 \) sign. In the range of parameters, the final \( \Delta Q \) is independent of \( t_0 \), i.e., how the voltage is applied. Here, we conclude that the MCD induces charge difference \( \Delta Q \) in the capacitor when flipping the magnetization from \( M \) to \( -M \).

The \( \Delta Q \) amplitude is sensitive to device parameters. We take the DC case for example. We adopt the following parameters by considering similar nano junctions(e.g., Ref.26), \( R = 10^9 \ \Omega, \ R_D = 10^9 \ \Omega, \ C = 10^{-20} \ F, \ D_M^x = 0.4, \) and \( V_0 = 1 \ \text{V} \) if not specified. (i) It is clear that \( \Delta Q = 0 \) without the diode \( (D_M^x = 0) \). As increasing \( D_M^x \), \( \Delta Q \) increases almost linearly (see Figure 2e). (ii) \( \Delta Q \) is an even function of \( V_0 \) and shows approximately a quadratic relation (see Figure 2d) because larger \( V_0 \) increases simultaneously the rectification effect (the \( V_D^2 \) term in Eq.1) and capacitor voltage. (iii) \( \Delta Q \) turns to zero for \( R \rightarrow 0 \) when the capacitor is shorted. \( \Delta Q \) also approaches zero if \( R \rightarrow \infty \) when there is nearly no current flow. Then it is natural to expect that \( \Delta Q \)
reaches a maximum at the intermediate range, as shown by Figure 2f. Therefore, we state that the magnetochiral charge correction is induced by the current flow and characterizes the nonequilibrium state and $\Delta Q$ diminishes if the chiral layer is rather metallic.

In experiments, a low-frequency ($1 \sim 1000$ Hz) AC bias was also applied such as $V_b(t) = V_0 \sin 2\pi ft$ and provides the same MR as the DC case. The RC circuit sets an upper-bound for the frequency $2\pi f \ll \frac{1}{RC}$. Given that $R = 10^9 \Omega$ and $C = 0.01 \sim 1 \times 10^{-18}$ F, $f \ll 1 \sim 100$ GHz holds in general experimental conditions. As shown in Figure 2c, $\Delta Q$ at peak positions appears to be the same as the DC case even for a large $f = 0.1$ GHz. Corresponding $\Delta Q$ dependence on $D_M^\chi$, $V_0$ or $R$ agrees quantitatively with the DC case.

**Tunneling barrier modulation.** The insulating chiral molecule acts as a tunneling barrier between two metallic leads. The current-driven charge accumulation can modulate the profile of the tunneling potential and thus, will sensitively change the resultant resistance.

Further, we demonstrate the high-order magnetoresistance by simulating the I-V curve. For simplicity, we consider tunneling through a one-dimensional potential barrier. It is natural to presume that the chiral molecule carries an intrinsic charge dipole $27,28$ characterizing the original potential barrier $E_0r$ across the molecule, where $E_0$ is the dipole field and $r$ is the position. When current flows, the driven charge accumulation $\Delta Q$ induces an extra field $\Delta Er$, as discussed above. We take the relation $\Delta E = \lambda_M V^2$, where $V$ is the bias and $\lambda_M = -\lambda_{-M}$ is an $M$-dependent coefficient, according to the results in Figure 2d. Therefore, the total barrier potential ($U$) is bias
dependent,

\[ U = U_0 + (E_0 + \lambda_M V^2) r, \]  

(4)

where \( U_0 \) is the barrier height without intrinsic and extrinsic dipoles. The tunneling current can be sensitively modified even for small potential changes. In addition, the intrinsic dipole field \( (E_0 r) \) is necessarily involved. Otherwise, potential barriers are symmetric for \( \pm M \) and lead to the same tunneling probability. As shown in Fig. 3b, we obtain I-V curves for \( \pm M \), which are qualitatively similar to experimental ones. Here, the MR reaches 85% for merely 10% induced dipole change \( (\lambda_M V^2 / E_0) \) at 1 V bias in the simulation. The MR increases as increasing the barrier width, as shown in Fig. 3c. The width dependence agrees with the experimental fact that MR increases by increasing molecule length \(^7, 13, 21, 29, 30\). Here, much lower conductance comes together with the wider barrier and thus is more sensitive to the potential modification. We point out that the MR magnitude is not necessarily constrained by the spin polarization in the lead. In theory, the high-order MR of CISS can approach 100% at large bias or long molecules.

3 Discussion

We show that MCD leads to magnetization-dependent nonequilibrium charge accumulation at both lead-molecule interfaces, which is distinct from the equilibrium polarization proposed in Ref. 22 recently. Subsequently, the resultant resistance changes between \( M \) and \(-M\) due to the modified tunneling barrier through the molecule. Recalling \( \Delta Q \propto V^2 \) (Figure 2d) and Eq. 1, we can expand the resistance \( R_{tot} \) to the leading order of \( \Delta Q \) and obtain approximately \( R_{tot} = R_0(1 + D_M^x V + \gamma_M V^2) \), where \( R_0 \) and \( \gamma_M = -\gamma_{-M} \) are coefficients. Then we get an I-V relation
for given $M$ in the small bias regime,

$$I = \frac{1}{R_0} (V - D_M^\chi V^2 - \gamma_M V^3).$$

(5)

Corresponding I-V curve displays approximate antisymmetry like the Figure 1c if the $V^3$ term is larger than $V^2$ term. This is possibly the case in experiments because charge accumulation may be significant at the metal-insulator interface. We note that the $V^2$ term is induced by the symmetry-breaking and does not necessarily vanish. Thus, CISS I-V curves may deviate from the ideal anti-symmetry, which is the case in many experiments.

As a highly nonlinear effect, CISS MR can provide a platform to study the fluctuation-dissipation theorem\textsuperscript{31–33}, which describes nonlinear responses beyond Onsager’s reciprocal theorem. Fluctuation theorems provide the relation between current-noise and high-order conductance\textsuperscript{34}. Experimental I-V curves are indeed noisy (e.g., see Refs.7, 11–13, 35). We propose that noise analysis on the experimental data may provide more insights into the nonlinear nature of CISS MR.

As shown above, the higher-order nature of CISS MR relies on the coexistence of MCD and capacitor. If the chiral layer turns to be metallic and shorts the capacitor, we anticipate that the CISS device turns into an ordinary MCD. Actually, recent experiments support this conclusion, where \textit{metallic} chiral crystals (CrNb$_3$S$_6$ and Nb(Ta)Si$_2$) were investigated\textsuperscript{36–38}. These chiral metals present electric magnetochiral anisotropy (see Figure 1d), although it was naively claimed to be CISS-type MR in Ref.37, 38. If the MCD turns weak in rectification, CISS MR will become weak too. As revealed in Ref. 20, MCD relies on SOC in leads and molecules. We propose that replacing heavy-metal leads with light metals (e.g., Al or Cu) in current CISS devices will significantly reduce
In open-circuit case ($R = \infty$), an AC bias can generate MCD-dependent charge accumulation in a CISS device. It is different from the low-frequency limit because $\omega \gg \frac{1}{RC}$ always holds here. We propose that such charge accumulation will lead to surface potential change which modulates surface-related properties. Beyond MR, our model can rationalize a seemingly unrelated experiment on the work function measurement \cite{21}. On a ferromagnetic thin film coated by chiral molecules, the Kelvin probe detects potential change by 100 mV when flipping the magnetization. The Kelvin probe has a capacitor setup with periodically modulating ($f = 75$ KHz in Ref. 21) electrode-surface distance, which is equivalent to an AC bias. We simulate CISS in the Kelvin probe by a capacitor-MCD circuit. As shown in Figure 3, the induced charge accumulation difference ($\Delta Q$) increases quickly with increasing frequency. Here, we use a larger capacitance ($C = 10^{-17}$ F) than that of Figure 2 to enhance the charging effect in calculations. It is natural to expect $\Delta Q = 0$ at $f = 0$ Hz because it represents a static equilibrium phase. Thus, we propose that the measured work function is a dynamic quantity and its frequency dependence should be examined in future experiments. Furthermore, the work function change can be correlated to the MR amplitude under similar experimental conditions because it characterizes the charge accumulation. In other words, the work function change is another characteristic feature of CISS like the MR.

Furthermore, we propose the recently discovered twisted van der Waals materials \cite{39-41} as a versatile platform for the CISS effect. In twisted bilayer graphene and twisted transition-metal dichalcogenides \cite{42-45}, the in-plane transport has been extensively studied for flat-band-driven phe-
Figure 3: **Simulated I-V curves of CISS and open-circuit charge accumulation.** (a) Opposite $M$ reshapes the tunneling potential barrier. The tunneling probability is simulated in a 1D lattice model where the potential change is quadratic to the bias voltage. (b) The calculated I-V curve for $\pm M$ calculated on a barrier of 65-lattice-site wide. (c) The magnetoresistance (MR) ratio increases as increasing the barrier width, which is in the unit of lattice sites, for the same dipole amplitude. It is consistent with the MR dependence on the molecule length in experiments. (d) The charge accumulation difference ($\Delta Q$) in an open circuit for the AC bias. It increases quickly as increasing frequency, which can be detected by the work function change in experiments.
Figure 4: **CISS platforms by twisted van der Waals layers sandwiched between vertical electrodes.** The electrodes can be either metals or metallic/semimetallic van der Waals materials. Blue arrows indicate the magnetic material. Magnetism can appear in one/both electrodes (a) and/or (b) in the twisted material. If the twisted layers are insulating, CISS-type magnetoresistance occurs. If it is more conducting, it acts as a magnetochiral diode. The role of SOC can be examined in different electrodes like graphite.
nomina such as the unconventional superconductivity, Mott insulating states, and Chern insulators. We conceive a two-terminal vertical device where two electrodes sandwich twisted bilayers or multiple layers, as illustrated in Figure 4. Very recently, it was proposed that flat bands significantly enhance the MCD effect in twisted materials\textsuperscript{46}. The MCD comes from the coexistence of magnetization (or magnetic field) and chirality. Since twisted layers are chiral, we introduce magnetism to either one electrode and/or twisted layers. Many van der Waals materials are magnetic, such as insulating CrI\textsubscript{3}\textsuperscript{47}, semiconducting MnBi\textsubscript{2}Te\textsubscript{4}\textsuperscript{48} and metallic Fe\textsubscript{3}GeTe\textsubscript{2}\textsuperscript{49}. Additionally, twisting between van der Waals electrodes can also play the role of chirality. The vertical resistance can be measured for the opposite magnetization and for opposite twist angles. If chiral layers are insulating, we anticipate CISS-like MR. If they are metallic, we expect MCD-like MR. In most the present CISS devices, a thin layer of gold is technically required to protect the ferromagnetic film. This gold layer results in difficulties to verify the influence of substrate SOC\textsuperscript{20,50}. In Figure 4, electrodes can also be made of van der Waals materials such as graphite with negligible SOC, which circumvents this problem as well as the complexity of metal-organic molecule junction. Therefore, van der Waals materials provide a versatile platform to investigate the relation of CISS with SOC, magnetism, and the chiral layer insulation. It would be more exciting to examine the influence of flat bands\textsuperscript{39,46}, correlation effects, and even superconductivity in CISS\textsuperscript{51}.

4 Summary

In summary, we reveal that CISS MR is a high-order effect emerging from the current-induced magnetochiral polarization. Our theory explains several significant experimental facts together: (i)
The CISS MR increases with the molecule length and has a giant magnitude up to the theoretical limit of 100%. (ii) Insulating chiral molecules exhibit giant, third-order (or even higher) MR while some metallic chiral solids show small, second-order MR, i.e., electric magnetochiral anisotropy. (iii) Chiral molecules modify the work function of a ferromagnetic substrate. Further, we made several predictions that can be examined in experiments: (i) The amplitude of MR will decrease if the chiral layer turns more metallic because metallicity suppresses the charge polarization. (ii) The CISS is sensitive to the electrode SOC. For example, substituting Al or Cu for Au electrodes will significantly reduce MR. (iii) The CISS-driven work function change is dynamical and increases with the measurement frequency. Furthermore, our work provides insights to design and understand novel nanodevices and generalize CISS to twisted van der Waals materials.

Our model is based on general symmetry principles and is independent of microscopic details, for example, the origin of SOC, phonon, or the correlation effect. We investigate the charge transport – the directly measurable quantity in CISS, instead of the spin polarization. But it does not necessarily exclude the existence of spin polarization, which can be measured by other techniques. Distinct from electric magnetochiral anisotropy, we propose that CISS MR is a higher-order effect derived from magnetism- and chirality-dependent nonequilibrium states. CISS refrains from the Onsager’s reciprocal relation because the charge accumulation breaks the microscopic reversibility.
Methods

The tunneling probability and I-V curves are simulated in a 1D tight-binding lattice with the quantum transport package Kwant\textsuperscript{52}. The nearest neighbor hopping is set to $t = 2 \text{ eV}$. For fixed bias $V$, the potential is calculated according to Eq. 4 and added to the 1D chain. The potential parameters (see Eq. 4 and Figure 3) are $U_0 = 3 \text{ eV}$, $E_0 = 0.05 \text{ eV}$ per site distance, $\lambda_M = \text{sign}(M) \ast 0.005 \text{ eV}^{-2}$ per site distance. The current ($I$) is obtained by integrating the transmission probability from zero to $V$. Figure 3b is calculated for a barrier of 65 sites wide. The MR ratios in Figure 3c are evaluated for different barrier widths at bias $1 \text{ V}$. Because the tunneling current is highly nonlinear to the barrier height and width, the I-V curves and MR-width relations can be non-monotonic in some cases. But their general trends are consistent in a large parameter range.

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