Spin-Dependent Electron Transmission Model for Chiral Molecules in Mesoscopic Devices

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Abstract

Various device-based experiments have indicated that electron transfer in certain chiral molecules may be spin-dependent, a phenomenon known as the Chiral Induced Spin Selectivity (CISS) effect. However, due to the complexity of these devices and a lack of theoretical understanding, it is not always clear to what extent the chiral character of the molecules actually contributes to the magnetic-field-dependent signals in these experiments. To address this issue, we report here an electron transmission model that evaluates the role of the CISS effect in two-terminal and multi-terminal linear-regime electron transport experiments. Our model reveals that for the CISS effect, the chirality-dependent spin transmission is accompanied by a spin-flip electron reflection process. Furthermore, we show that more than two terminals are required in order to probe the CISS effect in the linear regime. In addition, we propose two types of multi-terminal nonlocal transport measurements that can distinguish the CISS effect from other magnetic-field-dependent signals. Our model provides an effective tool to review and design CISS-related transport experiments, and to enlighten the mechanism of the CISS effect itself.
I. INTRODUCTION

Developments in the semiconductor industry have allowed integrated circuits to rapidly shrink in size, reaching the limit of conventional silicon-based electronics. One idea to go beyond this limit is to use the spin degree-of-freedom of electrons to store and process information (spintronics). A spintronic device usually contains two important components: a spin injector and a spin detector, through which electrical or optical signals and spin signals can be interconverted. Conventionally, this conversion is done with bulky solid-state materials, but the recently discovered Chiral Induced Spin Selectivity (CISS) effect suggests that certain chiral molecules or their assemblies are capable of generating spin signals as well. This effect describes that electrons acquire a spin polarization while being transmitted through certain chiral (helical) molecules. Notably, experimental observations of the CISS effect suggest its existence, but complete theoretical insight in its origin is still lacking. The CISS effect is thus not only relevant for spintronic applications, but also fundamentally interesting.

The CISS effect has been experimentally reported in chiral (helical) systems ranging from large biological units such as dsDNA to small molecules such as helicenes. Typically, these experiments can be categorized into either electron photoemission experiments or magnetotransport measurements. The latter, in particular, are usually based on solid-state devices, and are thus of great importance to the goal of realizing chiral-molecule-based spintronics. Important to realize, in such devices, the CISS-related signals may often be overshadowed by other spurious signals that arise from magnetic components of the devices. Therefore, it is essential to understand the exact role of chiral molecules in these devices, and to distinguish between the CISS-related signals and other magnetic-field-dependent signals. However, this has not been addressed, and an effective tool to perform such analyses is still missing.

We provide here a model that is based on the Landauer-Büttiker-type of analysis of linear-regime electron transmission and reflection. Unlike other theoretical works, our model is derived from symmetry theorems that hold for electrical conduction in general, and does not require any assumptions about the CISS effect on a molecular level. With this model, we quantitatively demonstrate how the CISS effect leads to spin injection and detection in linear-regime devices, and analyze whether typical two-terminal and four-terminal measurements...
are capable of detecting the CISS effect in the linear regime.

II. MODEL

We consider a solid-state device as a linear-regime circuit segment whose constituents are described by the following set of rules:

• A contact (pictured as a wavy line segment perpendicular to the current flow, see e.g. Fig. 1) is described as an electron reservoir with a well-defined chemical potential $\mu$, which determines the energy of the electrons that leave the reservoir. A reservoir absorbs all incoming electrons regardless of its energy or spin;

• A node (pictured as a circle, see later figures for four-terminal geometries) is a circuit constituent where chemical potentials for charge and spin are defined. It is described by two chemical potentials $\mu_\rightarrow$ and $\mu_\leftarrow$, one for each spin species with the arrows indicating the spin orientations. At a node a spin accumulation $\mu_s$ is defined ($\mu_s = \mu_\rightarrow - \mu_\leftarrow$). Inside a node the momentum of electrons is randomized, while the spin is preserved;

• A CISS molecule (pictured as a helix, color-coded and labeled for its chirality, see e.g. Fig. 1), a ferromagnet (a filled square, see e.g. Fig. 1), or a non-magnetic barrier (a shaded rectangle, see e.g. Fig. 2), is viewed as a circuit constituent with energy-conserving electron transmission and reflection. It is described by a set of (possibly spin-dependent) transmission and reflection probabilities;

• The above constituents are connected to each other via transport channels (pictured as line segments along the current flow, see e.g. Fig. 1). In such channels both the momentum and the spin of electrons are preserved.

With this set of constituents and rules, we first derive a key transport property of CISS molecules, and then introduce a matrix formalism to quantitatively describe linear-regime transport devices.

A. Reciprocity and spin-flip reflection by chiral molecules

In order to characterize the CISS effect without having to understand it on a molecular level, we look at universal rules that apply to any conductor in the linear regime, namely
the law of charge conservation and the reciprocity theorem.

The reciprocity theorem states that for a multi-terminal circuit segment in the linear regime, the measured conductance remains invariant when an exchange of voltage and current contacts is accompanied by a reversal of magnetic field $H$ and magnetization $M$ (of all magnetic components)$^{27,28}$ Mathematically we write

$$G_{ij,mn}(H, M) = G_{mn,ij}(-H, -M), \quad (1)$$

where $G_{ij,mn}$ is the four-terminal conductance measured using current contacts $i$ and $j$ and voltage contacts $m$ and $n$. In two-terminal measurements this theorem reduces to

$$G_{ij}(H, M) = G_{ij}(-H, -M), \quad (2)$$

meaning that the two-terminal conductance remains constant under magnetic field and magnetization reversal. This theorem emphasizes the universal symmetry independent of the microscopic nature of the transport between electrical contacts. It is valid for any linear-regime circuit segment regardless of the number of contacts, or the presence of inelastic scattering events$^{28}$

![Diagram](image)

**FIG. 1:** A two-terminal circuit segment with a $P$-type CISS molecule and a ferromagnet between contacts 1 and 2 (with chemical potentials $\mu_1$ and $\mu_2$). The notion $P$-type represents the chirality of the molecule, and indicates that it allows higher transmission for spins parallel to the electron momentum. (The opposite chirality allows higher transmission for spins anti-parallel to the electron momentum, and is denoted as $AP$-type.) The ferromagnet ($FM$) is assumed to allow higher transmission of spins parallel to its magnetization direction, which can be controlled to be either parallel or anti-parallel to the electron transport direction.

By applying the reciprocity theorem to a circuit segment containing CISS molecules, one can derive a special transport property of these molecules. For example, in the two-terminal circuit segment shown in Fig. 1, the reciprocity theorem requires that the two-terminal conductance remains unchanged when the magnetization direction of the ferromagnet is
reversed. Since the two-terminal conductance is proportional to the transmission probability $T_{12}$ between the two contacts (Landauer-Büttiker),\textsuperscript{29} this requirement translates to

$$T_{12}(\Rightarrow) = T_{12}(\Leftarrow),$$

where $\Rightarrow$ and $\Leftarrow$ indicate the magnetization directions of the ferromagnet. This requirement gives rise to a necessary spin-flip process associated with the CISS molecule, as described below.

For ease of illustration, we assume an ideal case, where both the ferromagnet and the CISS molecule allow a 100% transmission of the favored spin and a 100% reflection of the other, and the general validity of the conclusions is addressed in the supplementary information. We consider electron transport from contact 1 to contact 2 (see Fig. 1), and compare the two transmission probabilities $T_{12}(\Rightarrow)$ and $T_{12}(\Leftarrow)$. For $T_{12}(\Rightarrow)$, the $P$-type CISS molecule (favors spin parallel to electron momentum, see figure caption) allows the transmission of spin-right electrons, while it reflects spin-left electrons back to contact 1. At the same time, the ferromagnet is magnetized to also only allow the transmission of spin-right electrons. Therefore, all spin-right (and none of the spin-left) electrons can be transmitted to contact 2, giving $T_{12}(\Rightarrow) = 0.5$. As for $T_{12}(\Leftarrow)$, while the $P$-type CISS molecule still allows the transmission of spin-right electrons, the ferromagnet no longer does. It reflects the spin-right electrons towards the CISS molecule with their momentum anti-parallel to their spins. As a result, these electrons are reflected by the CISS molecule, and are confined between the CISS molecule and the ferromagnet. This situation gives $T_{12}(\Leftarrow) = 0$, which is not consistent with Eqn. 3. In order to satisfy Eqn. 3, i.e. have $T_{12}(\Leftarrow) = 0.5$, a spin-flip process has to take place for the spin-right electrons, so that they can be transmitted to contact 2 through the ferromagnet. Such a process does not exist for the ideal and exactly aligned ferromagnet. Therefore, a spin-flip electron reflection process must exist for the CISS molecule. Further analysis (see supplementary information) shows that such a spin-flip reflection process completely meets the broader restrictions from Eqn. 2. In addition, the conclusion that a spin-flip reflection process must exist is valid for general cases where the ferromagnet and the CISS molecule are not ideal (see supplementary information).

In these derivations the only assumption regarding the CISS molecule is that it allows higher transmission of one spin than the other, which is a conceptual description of the CISS effect itself. Therefore, the spin-flip reflection process has to be regarded as an inherent
property of the CISS effect in a linear-regime transport system, and this is guaranteed by the universal symmetry theorems of electrical conduction.27,28

B. Matrix formalism and barrier-CISS center-barrier (BCB) model for CISS molecules

We use matrices to quantitatively describe the spin-dependent transmission and reflection probabilities of CISS molecules and other circuit constituents, as shown in Fig. 2. At the top
of the figure, the general form of these matrices is introduced. Matrix elements $t_{\alpha\beta}$ (or $r_{\alpha\beta}$),
where $\alpha$ and $\beta$ is either left ($\leftarrow$) or right ($\rightarrow$), represent the probability of a spin-$\alpha$ electron being transmitted (or reflected) as a spin-$\beta$ electron, and $\alpha \neq \beta$ indicates a spin-flip process. Here $0 \leq t_{\alpha\beta}, r_{\alpha\beta} \leq 1$, and the spin orientations are chosen to be either parallel or anti-parallel to the electron momentum in later discussions. Next, the transmission and reflection matrices of a non-magnetic barrier are given. These matrices are spin-independent and are fully determined by a transmission probability $t$ ($0 \leq t \leq 1$), which depends on the material and dimensions of the barrier. Here we use the term barrier, but it refers to any circuit constituent with spin-independent electron transmission and reflection. In the third row, we show the transmission and reflection matrices of a ferromagnet. These matrices are spin-dependent, and are determined by the polarization $P_{FM}$ ($0 < |P_{FM}| \leq 1$) of the ferromagnet.

Finally, for $P$-type and $AP$-type CISS molecules, we show here an ideal case where all the matrix elements are either 1 or 0. The non-zero off-diagonal terms in the reflection matrices represent the characteristic spin-flip reflection. These ideal CISS molecules are later referred to as CISS centers, and will be generalized for more realistic situations.

In accordance with the matrix formalism, we use row vectors $\boldsymbol{\mu} = (\mu_{\rightarrow}, \mu_{\leftarrow})$ to describe chemical potentials, and row vectors $\mathbf{I} = (I_{\rightarrow}, I_{\leftarrow})$ to describe currents, where each vector element describes the contribution from one spin component (indicated by arrow).

![Diagram](image.png)

FIG. 3: A generalized Barrier-CISS Center-Barrier (BCB) model for $P$-type CISS molecules. The ideal, 100%-spin-selective CISS Center in the middle introduces the directional spin transmission in a CISS molecule, while the two identical non-magnetic barriers (with transmission probability $t$) contribute the non-ideal electron transmission and reflection behavior. The overall transmission and reflection matrices of the entire BCB module are fully determined by $t$, and have all elements taking finite values between 0 and 1.

A non-ideal CISS molecule with $C_2$ symmetry (two-fold rotational symmetry with axis perpendicular to the electron transport path) can be modeled by a linear arrangement of
two identical barriers sandwiching an ideal CISS center, as shown in Fig. 3 (only the \(P\)-type is shown). In this \textit{Barrier-CISS Center-Barrier (BCB) model} we consider that all spin-dependent linear-regime transport properties of these molecules exclusively originate from an ideal CISS center inside the molecules, and the overall spin-dependency is limited by the multiple spin-independent transmissions and reflections at other parts (non-magnetic barriers) of the molecule. Therefore, the barrier transmission probability \(t\) \((0 < t \leq 1)\) fully determines the transmission and reflection matrices of the entire BCB molecule, and consequently determines the spin-related properties of the molecule. The use of an identical barrier on each side of the CISS center is to address the \(C_2\) symmetry. However, we stress that not all CISS molecules have this symmetry, and the BCB model is still a simplified picture. The model can be further generalized by removing the restriction of the CISS center being ideal, and this case is discussed in the supplementary information. Despite being a simplified picture, the BCB model captures all qualitative behaviors of a non-ideal CISS molecule, and at the same time keeps quantitative analyses simple. Therefore, we further only discuss the case of \textit{BCB molecules}, instead of the more generalized \textit{CISS molecules}.

### III. DISCUSSIONS

In this section, we use different approaches to separately analyze two-terminal and multi-terminal circuit geometries. In two-terminal geometries, we evaluate the conductance of the circuit segment by calculating the electron transmission probability \(T_{12}\) between the two contacts. In contrast, for multi-terminal cases we take a circuit-theory approach to evaluate the spin accumulation \(\mu_s\) at the nodes. A major difference between the two approaches is the inclusion of the node in multi-terminal geometries. A node emits electrons to all directions equally, so it can be considered as a source of electron back-scattering. Notably, adding a node to a near-ideal electron transport channel (with transmission probability close to 1) significantly alters its electron transmission probability. This may make our two approaches look inconsistent with each other. However, this is not the case because we only address non-ideal circuit segments where electron back-scattering (reflection) already exists due to other circuit constituents (CISS molecules, ferromagnets, or non-magnetic barriers). Note that even when we discuss the use of ideal CISS molecules or ideal ferromagnets, the entire circuit segment is non-ideal due to the reflection of the rejected spins. Additionally, we
emphasize that for both two-terminal and multi-terminal geometries, our model does not account for effects originating from quantum phase coherence in the electron transport.

In these discussions we consider only the $P$-type BCB molecule, and we use expressions $T_{R,L}^P$ and $R_{R,L}^P$ to describe transmission and reflection matrices of the entire BCB module, where the subscripts consider electron flow directions. The derivations of these matrices can be found in the supplementary information.

A. Two-terminal geometries

We discuss here two geometries that are relevant for two-terminal magnetoresistance measurements.

![FM-BCB geometry](image)

**FIG. 4: A FM-BCB geometry where a ferromagnet and a BCB molecule are connected in series in a two-terminal circuit segment. The magnetization reversal of the ferromagnet does not change the two-terminal conductance.**

The first is a $FM$-$BCB$ geometry, as shown in Fig. 4. It simulates a common type of experiment where a layer of chiral molecules is sandwiched between a ferromagnetic layer and a normal metal contact. The other side of the ferromagnetic layer is also connected to a normal metal contact (experimentally this may be a wire that connects the sample with the measurement instrument). Due to the spin-dependent transmission of the chiral molecules and the ferromagnet, one might expect a change of the two-terminal conductance once the magnetization of the ferromagnet is reversed. However, this change is not allowed by the reciprocity theorem (Eqn. 2), which can also be confirmed with our model, as shown below.

In order to illustrate this, we calculate the electron transmission probabilities between the two contacts for opposite ferromagnet magnetization directions, $T_{12}^{FM-BCB}(\rightarrow)$ and $T_{12}^{FM-BCB}(\leftarrow)$, where the arrows indicate the magnetization directions.

For the magnetization direction to the right (⇒), we first derive the transmission and reflection matrices with the combined contribution from the ferromagnet and the BCB...
molecule

\[
T_{12}^{FM-BCB}(\Rightarrow) = T_{FM}(\Rightarrow) \cdot \left( \mathbb{1} + R_R^P \cdot R_{FM}(\Rightarrow) \right) + \left( R_R^P \cdot R_{FM}(\Rightarrow) \right)^2 + \left( R_R^P \cdot R_{FM}(\Rightarrow) \right)^3 + \cdots \cdot T_R^P \cdot \left( \mathbb{1} - R_R^P \cdot R_{FM}(\Rightarrow) \right)^{-1} \cdot T_R^P
\]

(4a)

\[
R_{11}^{FM-BCB}(\Rightarrow) = R_{FM}(\Rightarrow) + T_{FM}(\Rightarrow) \cdot R_R^P \cdot \left( \mathbb{1} - R_R^P \cdot R_{FM}(\Rightarrow) \right)^{-1} \cdot T_{FM}(\Rightarrow),
\]

(4b)

where the \( \mathbb{1} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \) is the identity matrix. The addition of the multiple reflection terms is due to the multiple reflections between the ferromagnet and the BCB molecule. Next, we include the contribution from the contacts and derive the transmission and reflection probabilities between the contacts

\[
T_{12}^{FM-BCB}(\Rightarrow) = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} T_{12}^{FM-BCB}(\Rightarrow) \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

(5a)

\[
R_{11}^{FM-BCB}(\Rightarrow) = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} R_{11}^{FM-BCB}(\Rightarrow) \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

(5b)

where the row vector \( \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} \) describes the input current from contact 1 with equal spin-right and spin-left contributions, and the column vector \( \begin{pmatrix} 1 \\ 1 \end{pmatrix} \) describes the absorption (sum) of both spins into contact 2.

For the opposite magnetization direction (\( \Leftarrow \)), we change the magnetization-dependent terms in Eqn. 4 and Eqn. 5 accordingly. Detailed calculations (see supplementary information) give

\[
T_{12}^{FM-BCB}(\Rightarrow) \equiv T_{12}^{FM-BCB}(\Leftarrow)
\]

(6)
for all BCB transmission probabilities $t$ and all ferromagnet polarizations $P_{FM}$. Therefore, it is not possible to detect any variation of two-terminal conductance in this geometry by switching the magnetization direction of the ferromagnet. In the supplementary information we show that it is also not possible to detect any variation of two-terminal conductance by reversing the current, and that the above conclusions also hold for the more generalized CISS model. These conclusions agree with earlier reports on general voltage-based detections of current-induced spin signals.$^{31}$

![Spin Valve geometry](image)

FIG. 5: A Spin Valve geometry with a BCB molecule placed in between two ferromagnets.

Unlike a conventional spin valve, in this geometry the magnetization reversal of one ferromagnet does not change the two-terminal conductance, due to the presence of the BCB molecule.

The second geometry, as shown in Fig. 5 contains two ferromagnets, and is similar to a conventional spin valve. In a spin valve, it is known that when the magnetization direction of one ferromagnet is reversed, the two-terminal conductance changes.$^{30}$ This does not violate the reciprocity theorem since switching one ferromagnet does not reverse all magnetizations of the entire circuit segment. In contrast, in the geometry shown in Fig. 5 due to the presence of the spin-flip electron reflection, our analysis (see supplementary information) shows that the magnetization reversal of one ferromagnet does not affect the two-terminal conductance. As a result, this geometry is not able to quantitatively measure the CISS effect. We emphasize that this absence of the spin valve behavior is unique for the BCB model, which contains an ideal CISS Center. In the supplementary information we show that a further-generalized CISS model regains the spin valve behavior. Nevertheless, one cannot experimentally distinguish whether the spin valve behavior originates from the CISS effect or a normal non-magnetic barrier, and therefore cannot draw any conclusion about the CISS effect. In general, it is not possible to measure the CISS effect in the linear regime using two-terminal experiments.
B. Four-terminal geometries and experimental designs

Four-terminal measurements allow one to completely separate spin-related signals from charge-related signals, and therefore also allow the detection of spin accumulation created by the CISS effect. Here we analyze two geometries that are relevant for such measurements. In the first geometry, we use a node connected to BCB molecules to illustrate how spin injection and detection can occur without using magnetic materials (Fig. 6). In the second geometry, we use two nodes to decouple a BCB molecule from electrical contacts, and illustrate the spin-charge conversion property of the molecule (Fig. 7). In addition, we propose device designs that resemble these two geometries and discuss possible experimental outcomes (Fig. 8).

FIG. 6: a). A four-terminal geometry involving a node. Two of the contacts contain BCB molecules, and the other two are coupled to the node via tunnel barriers (with transmission probability \( t_B \)). The node can be characterized by a spin-dependent chemical potential vector \( \mu_{\text{node}} = (\mu_{\text{node} \rightarrow}, \mu_{\text{node} \leftarrow}) \), and each of the four contacts is characterized by a spin-independent chemical potential \( \mu_i \), with \( i = 1, 2, 3, 4 \). b). Calculated ratio between four-terminal and two-terminal resistances for this geometry, plotted as a function of \( t \) (transmission probability of the barriers in BCB molecules) for three \( t_B \) (transmission probability of the barriers at the contacts) values.

Fig. 6a) shows a geometry where a node is connected to four contacts. Two of the contacts contain BCB molecules, and the other two contain non-magnetic (tunnel) barriers. We consider an experiment where contacts 1 and 2 are used for current injection and contacts
3 and 4 are used for voltage detection. In terms of spin injection, we first assume that the voltage contacts 3 and 4 are weakly coupled to the node, and do not contribute to the spin accumulation in the node. This means that the chemical potentials of contacts 1 and 2 fully determine the spin-dependent chemical potential (row vector) of the node

\[ \mu_{\text{node}} = (\mu_{\text{node} \rightarrow}, \mu_{\text{node} \leftarrow}) \].

We also assume \( \mu_2 = 0 \) for convenience since only the chemical potential difference between the two contacts is relevant. Under these assumptions, the node receives electrons from contact 1, and emits electrons to contacts 1 and 2. Therefore, the incoming current into the node is

\[ I_{\text{in}} = \frac{G}{e} \left( \mu_1 (1, 1)^T P_R \right), \tag{7} \]

and the outgoing current from the node is

\[ I_{\text{out}} = \frac{G}{e} \left( \mu_{\text{node}} (1 - R_L^P) + \mu_{\text{node}} (1 - t_B) \right), \tag{8} \]

where \( G = N e^2 / h \) is the \( N \)-channel, one-spin conductance of the channels connecting the node to each of the contacts, and \( t_B (0 \leq t_B \leq 1) \) is the reflection probability of the tunnel barrier between the node and contact 2 (not the barrier in a BCB module). Due to the spin-preserving nature of the node, at steady state the incoming current is equal to the outgoing current (for both spin components), or \( I_{\text{in}} = I_{\text{out}} \). From this relation we derive

\[ \mu_{\text{node}} = \mu_1 (1, 1)^T P_R \left( (1 + t_B) I - R_L^P \right)^{-1}, \tag{9} \]

where \( t_B = 1 - r_B \) is the transmission probability of the tunnel barrier. Next, we derive the spin accumulation in the node

\[ \mu_s = \mu_{\text{node} \rightarrow} - \mu_{\text{node} \leftarrow} = k_{\text{inj}} \mu_1, \tag{10} \]

where

\[ k_{\text{inj}} = (1, 1)^T P_R \left( (1 + t_B) I - R_L^P \right)^{-1} \begin{pmatrix} 1 \\ -1 \end{pmatrix}, \tag{11} \]

with \( 0 < k_{\text{inj}} \leq \frac{1}{4} \),

is the spin injection coefficient for these current contacts. This expression shows that the spin accumulation in the node depends linearly on the chemical potential difference between
the current contacts, and the coefficient $k_{\text{inj}}$ is determined by both the BCB molecule (with parameter $t$) and the tunnel barrier connected to contact 2 (with parameter $t_B$).

With regard to spin detection, we discuss whether the established spin accumulation in the node $\mu_s$ can lead to a chemical potential difference (and thus a charge voltage) between the weakly coupled voltage contacts 3 and 4. A contact cannot distinguish between the two spin components, therefore only the charge current (sum of both spins) is relevant. At steady state, there is no net charge current at any of the voltage contacts,

$$I_3 = \frac{G}{e} \left( \mu_3 (1,1) (1 - r_B) - \mu_{\text{node}} t_B \right) \begin{pmatrix} 1 \\ 1 \end{pmatrix} = 0,$$  \hspace{1cm} (12a)$$

$$I_4 = \frac{G}{e} \left( \mu_4 (1,1) (I - R_L^P) - \mu_{\text{node}} T_R^P \right) \begin{pmatrix} 1 \\ 1 \end{pmatrix} = 0,$$  \hspace{1cm} (12b)$$

which gives

$$\mu_4 - \mu_3 = k_{\text{det}} \mu_s,$$  \hspace{1cm} (13)$$

where

$$k_{\text{det}} = \frac{1}{2} \begin{pmatrix} 1, -1 \end{pmatrix} T_R^P \begin{pmatrix} 1 \\ 1 \end{pmatrix},$$  \hspace{1cm} (14)$$

with $0 < k_{\text{det}} \leq \frac{1}{2}$, is the spin detection coefficient for these voltage contacts. This expression shows that the chemical potential difference between the two voltage contacts depends linearly on the spin accumulation in the node, and the coefficient $k_{\text{det}}$ is exclusively determined by the BCB molecule (with parameter $t$).

Combining Eqn. 10 and Eqn. 13 we obtain

$$R_{4T} = \frac{\mu_4 - \mu_3}{\mu_1 - \mu_2} = k_{\text{inj}} k_{\text{det}},$$  \hspace{1cm} (15)$$

where $R_{4T}$ is the four-terminal resistance (measured using contacts 3 and 4 as voltage contacts, while using contacts 1 and 2 as current contacts), and $R_{2T}$ is the two-terminal resistance (measured using contacts 1 and 2 as both voltage and current contacts). This ratio is
determined by both the BCB molecule (with parameter $t$) and the tunnel barrier connected to contact 2 (with parameter $t_B$), and can be experimentally measured to quantitatively characterize the CISS effect.

As an example, for $t = t_B = 0.5$, we have $k_{\text{inj}} \approx 0.11$, $k_{\text{det}} \approx 0.17$, and $R_{4T}/R_{2T} \approx 0.02$. In Fig. 6(b) we plot $R_{4T}/R_{2T}$ as a function of $t$ for three different $t_B$ values. Similar plots for $k_{\text{inj}}$ and $k_{\text{det}}$ are shown in the supplementary information.

The above results show that it is possible to inject and detect a spin accumulation in a node using only BCB molecules and non-magnetic (tunnel) barriers, and these processes can be quantitatively described by the injection and detection coefficients. We stress that the signs of the injection and detection coefficients depend on the type (chirality) of the BCB molecule and the position of the molecule with respect to the node. Switching the molecule from $P$-type to $AP$-type leads to a sign change of the injection or detection coefficient. The same happens if the node is connected to the opposite side of the BCB molecule. For example, in Fig. 6(a), contact 4 is connected to a BCB molecule to its right-hand side, and the node is connected to its left-hand side. If their positions are switched, i.e. having contact 4 connected to the left-hand side of the molecule, and the node connected to the right-hand side, the detection coefficient changes sign. This sign change can be experimentally measured as a signature of the CISS effect.

Fig. 7 shows a geometry where a BCB molecule is between two nodes $A$ and $B$, and is decoupled from the contacts. The nodes themselves are connected to contacts in a similar fashion as in the previous geometry. In node $A$, we consider a chemical potential vector $\mu_A$ and consequently a spin accumulation $\mu_{sA}$, which are fully determined by the current contacts 1 and 2. In node $B$, we consider weakly coupled voltage contacts 3 and 4, such that its chemical potential vector $\mu_B$, and hence its spin accumulation $\mu_{sB}$, are fully determined.
by $\mu_A$. At steady state, there is no net charge or spin current in node $B$, which leads to

$$\mu_B = \mu_A T_R^P (I - R_L^P)^{-1}. \quad (16)$$

Note that here the matrices only refer to the molecule between the two nodes. For BCB molecules, this expression always gives $\mu_{sB} = 0$, but for a more generalized CISS molecule (as described in the supplementary information), this expression can give $\mu_{sB} \neq 0$. This shows that a spin accumulation at one side of a CISS molecule can generate a spin accumulation at the other side of the molecule. Most importantly, for both the BCB model and the more generalized model, Eqn. [16] predicts that a spin accumulation difference across a CISS molecule creates a charge voltage across the molecule, and *vice versa* (spin-charge conversion via a CISS molecule). Mathematically written, the expression always provides $\mu_{nA} \neq \mu_{nB}$ when $\mu_{sA} \neq \mu_{sB}$, and $\mu_{sA} \neq \mu_{sB}$ when $\mu_{nA} \neq \mu_{nB}$, where $\mu_{nA}$ (or $\mu_{nB}$) is the average chemical potential of the two spin components in node $A$ (or in node $B$). A more detailed description of this geometry can be found in the supplementary information.

![Diagram](image)

**FIG. 8: Nonlocal device designs with CISS molecules adsorbed on graphene.** a). A device where electrons travel through CISS molecules. All contacts are non-magnetic and are numbered in agreement with Fig. 6a). A variation of this device can be achieved by replacing contact 1 with a ferromagnet, see inset. b). A device where electrons travel in proximity to CISS molecules. A ferromagnetic contact 2 is used for spin injection, but one can also use only non-magnetic contacts, as in Fig. 7.

Fig. 8 shows two types of nonlocal devices that resemble the two geometries introduced above. We realize the node function with graphene, chosen for its long spin lifetime and long spin diffusion length. The first type, as shown in Fig. 8a), represents the geometry in Fig. 6a), where spin injection and detection are both achieved using CISS molecules. A current $I_{inj}$ is injected
from contact 1 through CISS molecules into graphene, then driven out to a normal metal contact 2. This current induces a spin accumulation in the graphene layer underneath the current contacts, which then diffuses to the voltage contacts. The voltage contacts then pick up a charge voltage $V_{\text{det}}$ in a similar fashion as explained in Fig. 6a). With this, the nonlocal resistance can be determined $R_{\text{nl}} = V_{\text{det}}/I_{\text{inj}}$. Further, we can derive (see supplementary information for details)

$$R_{\text{nl}} = -\frac{1}{2}k_{\text{inj}}k_{\text{det}}R_{\text{inj}}e^{-\frac{d}{\lambda_s}},$$  \hspace{0em} (17)

where $R_{\text{inj}}$ is the resistance measured between the current contacts 1 and 2, $d$ is the distance between contacts 1 and 4, and $\lambda_s$ is the spin diffusion length of graphene. It is assumed here that the spacing between the current contacts (1 and 2) and the spacing between the voltage contacts (3 and 4) are both much smaller than $\lambda_s$. The factor 1/2 arises from the fact that the spin diffusion is symmetric in both directions, but the spin detector is located only on one side of the spin injector. The minus sign comes from the fact that the injection and the detection contacts are on the same side of the graphene channel (both on top), unlike the example in Fig. 6a) (one on the left and the other on the right).

A variation of this device is obtained by replacing contact 1 (together with the CISS molecules underneath it) with a ferromagnet, as shown in the inset of Fig. 8a). This variation allows one to control the sign of $R_{\text{nl}}$ by controlling the magnetization direction of the ferromagnet, which should be aligned parallel or anti-parallel to the helical (chiral) axis of the CISS molecules (out-of-plane). The nonlocal resistance is therefore

$$R_{\text{nl}}(\uparrow) = -R_{\text{nl}}(\downarrow) = \frac{1}{2}P_{\text{FM}}k_{\text{det}}R_{\lambda}e^{-\frac{d}{\lambda_s}},$$  \hspace{0em} (18)

where the arrows indicate the magnetization directions, $P_{\text{FM}}$ is the polarization of the ferromagnet, and $R_{\lambda}$ is the spin resistance of graphene (see supplementary information for more details). In this device, the reversal of the magnetization direction of the ferromagnet leads to a sign change of the nonlocal resistance. In experimental conditions, this nonlocal resistance change $\Delta R_{\text{nl}} = R_{\text{nl}}(\uparrow) - R_{\text{nl}}(\downarrow)$ can reach tens of Ohms (Ω), and is easily detectable.

The second type of device is depicted in Panel b). It is a variation of the geometry in Fig. 7, where contact 2 is replaced by a ferromagnet. In this device, instead of traveling through the CISS molecules, the electrons travel through the graphene channel underneath the molecules. It is assumed that due to the proximity of the CISS molecules, the electrons in
graphene also experience a (weaker) CISS effect. Whether this assumption is valid remains to be proven. The nonlocal signals produced by this device are derived in the supplementary information.

IV. CONCLUSION

In summary, we demonstrated that a spin-flip electron reflection process is inherent to the chiral induced spin selectivity (CISS) effect in linear-regime electron transport. Furthermore, we developed a set of spin-dependent electron transmission and reflection matrices and a generalized Barrier-CISS Center-Barrier (BCB) model to quantitatively describe the CISS effect in mesoscopic devices. Based on this formalism, we demonstrated that more than two terminals are needed in order to probe the CISS effect in linear-regime transport experiments. Moreover, we also showed several ways of injecting and detecting spins using CISS molecules, and demonstrated that CISS molecules can give rise to spin-charge conversion. In addition, we proposed two types of graphene-based nonlocal devices which can be used to directly measure the CISS effect in the linear regime.

We stress again that the above discussions and proposed devices are all based on linear-regime electron transport. Therefore, our conclusions cannot exclude the two-terminal detection of the CISS effect in the non-linear regime. However, the spin signals in non-linear-regime measurements should approach zero as the two-terminal bias approaches zero (entering the linear regime), and the mechanism that may contribute to such signals has to be different from spin-dependent electron transmission and reflection. A recent work shows that the CISS effect in electron photoemission experiments (three-terminal) can be explained by losses due to spin-dependent electron absorption in chiral molecules, but whether a similar process can lead to the detection of the CISS effect in non-linear two-terminal measurements remains to be investigated. Nevertheless, our model captures the fundamental role of the CISS effect in linear-regime mesoscopic devices. In addition, although our model does not assume any microscopic electron transport mechanism inside CISS molecules, the effects predicted by our model (for example the spin-flip reflection and spin-charge conversion) resemble spin-orbit effects. A new type of spin-orbit coupling was recently predicted for one-dimensional screw dislocations in semiconductor crystals, which has a one-dimensional helical effective electric field. This type of spin-orbit coupling can lead to an enhanced spin
lifetime for electrons traveling along the helical axis. Future theoretical work should study whether similar effects exist in chiral or helical molecules.

In general, our model helps to analyze and understand device-based CISS experiments without having to understand the CISS effect on a molecular level. It provides a guideline for future reviewing and designing of CISS-based mesoscopic spintronic devices.

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1 S. Wolf, D. Awschalom, R. Buhrman, J. Daughton, S. Von Molnar, M. Roukes, A. Y. Chtchelkanova, and D. Treger, Science 294, 1488 (2001).
2 R. Naaman and D. H. Waldeck, The Journal of Physical Chemistry Letters 3, 2178 (2012).
3 R. Naaman and D. H. Waldeck, Annual Review of Physical Chemistry 66, 263 (2015).
4 B. Göhler, V. Hamelbeck, T. Z. Markus, M. Kettner, G. F. Hanne, Z. Vager, R. Naaman, and H. Zacharias, Science 331, 894 (2011).
5 Z. Xie, T. Z. Markus, S. R. Cohen, Z. Vager, R. Gutierrez, and R. Naaman, Nano Letters 11, 4652 (2011).
6 V. Kiran, S. P. Mathew, S. R. Cohen, I. Hernández Delgado, J. Lacour, and R. Naaman, Advanced Materials 28, 1957 (2016).
7 M. Kettner, V. V. Maslyuk, D. Nürenberg, J. Seibel, R. Gutierrez, G. Cuniberti, K.-H. Ernst, and H. Zacharias, The Journal of Physical Chemistry Letters 9, 2025 (2018).
8 K. Ray, S. P. Ananthavel, D. H. Waldeck, and R. Naaman, Science 283, 814 (1999).
9 I. Carmeli, V. Skakalova, R. Naaman, and Z. Vager, Angewandte Chemie International Edition 41, 761 (2002).
10 S. G. Ray, S. S. Daube, G. Leitus, Z. Vager, and R. Naaman, Physical Review Letters 96, 036101 (2006).
11 D. Mishra, T. Z. Markus, R. Naaman, M. Kettner, B. Göhler, H. Zacharias, N. Friedman, M. Sheves, and C. Fontanesi, Proceedings of the National Academy of Sciences 110, 14872 (2013).
12 M. Á. Niño, I. A. Kowalik, F. J. Luque, D. Arvanitis, R. Miranda, and J. J. de Miguel, Advanced Materials 26, 7474 (2014).
13 M. Kettner, B. Göhler, H. Zacharias, D. Mishra, V. Kiran, R. Naaman, C. Fontanesi, D. H. Waldeck, S. Sek, J. Pawłowski, et al., The Journal of Physical Chemistry C 119, 14542 (2015).
14 K. Michaeli, V. Varade, R. Naaman, and D. H. Waldeck, Journal of Physics: Condensed Matter 29, 103002 (2017).
15 O. B. Dor, S. Yochelis, S. P. Mathew, R. Naaman, and Y. Paltiel, Nature Communications 4, 2256 (2013).
16 P. C. Mondal, N. Kantor-Uriel, S. P. Mathew, F. Tassinari, C. Fontanesi, and R. Naaman, Advanced Materials 27, 1924 (2015).
17 V. Varade, T. Markus, K. Vankayala, N. Friedman, M. Sheves, D. H. Waldeck, and R. Naaman, Physical Chemistry Chemical Physics 20, 1091 (2018).
18 B. P. Bloom, V. Kiran, V. Varade, R. Naaman, and D. H. Waldeck, Nano Letters 16, 4583 (2016).
19 S. P. Mathew, P. C. Mondal, H. Moshe, Y. Mastai, and R. Naaman, Applied Physics Letters 105, 242408 (2014).
20 K. M. Alam and S. Pramanik, Advanced Functional Materials 25, 3210 (2015).
21 R. Gutierrez, E. Díaz, R. Naaman, and G. Cuniberti, Physical Review B 85, 081404 (2012).
22 S. Yeganeh, M. A. Ratner, E. Medina, and V. Mujica, The Journal of Chemical Physics 131, 014707 (2009).
23 J. Gersten, K. Kaasbjerg, and A. Nitzan, The Journal of Chemical Physics 139, 114111 (2013).
24 E. Medina, L. A. González-Arraga, D. Finkelstein-Shapiro, B. Berche, and V. Mujica, The Journal of Chemical Physics 142, 194308 (2015).
25 K. Michaeli and R. Naaman, arXiv 1512.03435 (2015).
26 V. V. Maslyuk, R. Gutierrez, A. Dianat, V. Mujica, and G. Cuniberti, The Journal of Physical Chemistry Letters 9, 5453 (2018).
27 M. Büttiker, Physical Review Letters 57, 1761 (1986).
28 M. Büttiker, IBM Journal of Research and Development 32, 317 (1988).
29 M. Büttiker, Y. Imry, R. Landauer, and S. Pinhas, Physical Review B 31, 6207 (1985).
30 G. A. Prinz, Science 282, 1660 (1998).
31 I. Adagideli, G. E. W. Bauer, and B. I. Halperin, Physical Review Letters 97, 256601 (2006).
32 F. J. Jedema, A. T. Filip, and B. J. van Wees, Nature 410, 345 (2001).
33 M. Drögeler, C. Franzen, F. Volmer, T. Pohlmann, L. Banszerus, M. Wolter, K. Watanabe, T. Taniguchi, C. Stampfer, and B. Beschoten, Nano Letters 16, 3533 (2016).
34 J. Ingla-Aynés, M. H. D. Guimarães, R. J. Meijerink, P. J. Zomer, and B. J. van Wees, Physical Review B 92, 201410 (2015).
35 M. Gurram, S. Omar, S. Zihlmann, P. Makk, Q. Li, Y. Zhang, C. Schönenberger, and B. J. van Wees, Physical Review B 97, 045411 (2018).
36 D. Nürenberg and H. Zacharias, arXiv 1808.00269 (2018).
37 L. Hu, H. Huang, Z. Wang, W. Jiang, X. Ni, Y. Zhou, V. Zielasek, M. Lagally, B. Huang, and F. Liu, Physical Review Letters 121, 066401 (2018).
Supplementary Information: Spin-Dependent Electron Transmission Model for Chiral Molecules in Mesoscopic Devices

I. GENERAL VALIDITY OF THE SPIN-FLIP REFLECTION PROCESS

In the main text we stated that a spin-flip electron reflection process has to exist in order for spin-dependent transmission through CISS molecules to be allowed by the reciprocity theorem. Mathematically, this statement means that at least one off-diagonal term in the reflection matrices of CISS molecules has to be non-zero. Now we prove the general validity of our statement by limiting all off-diagonal terms of the reflection matrices to zero, and derive violations against the description of the CISS effect.

First of all, we write the general form of the transmission matrix of a CISS molecule

\[
T_{CISS} = \begin{pmatrix} a & b \\ c & d \end{pmatrix}, \text{ with } 0 \leq a, b, c, d \leq 1.
\]

For electrons traveling towards the CISS molecule, each spin component can either be transmitted (with or without spin-flip) or reflected, the sum of these probabilities is therefore unity. This can be written in matrix form

\[
(T_{CISS} + R_{CISS}) \begin{pmatrix} 1 \\ 1 \end{pmatrix} = \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

We assume that the spin-flip reflection process does not exist, i.e. if the off-diagonal terms of \( R_{CISS} \) are all zero, the above expression thus gives the only solution to \( R_{CISS} \)

\[
R_{CISS} = \begin{pmatrix} 1 - a - b & 0 \\ 0 & 1 - c - d \end{pmatrix}
\]

In addition, we adopt transmission and reflection matrices of a ferromagnet from Fig. 2.

Next, we consider that the CISS effect exists in the linear regime. According to the description of CISS effect, this means that with an input of spin non-polarized electrons, the CISS molecule gives a spin-polarized transmission output. Here we do not make assumptions about the chirality of the molecule or the electron flow direction, so that our conclusions hold for the most general situations. Therefore, we do not assume the sign of the polarization of
the transmission output, and write

\[
\begin{pmatrix} 1, 1 \end{pmatrix} T_{CISS} \begin{pmatrix} 1 \\ -1 \end{pmatrix} = (a + c) - (b + d) \neq 0,
\]

(S1)

where the row vector on the left indicates the spin non-polarized input current, and the column vector on the right calculates the difference between the two spin components in the output current.

We illustrate the discrepancy between the assumption of not having any spin-flip reflection process and the conceptual description of the CISS effect (Eqn. S1) by applying the reciprocity theorem to the circuit segment shown in Fig. 1 of the main text. For this circuit segment, we calculate the total transmission matrix accounting for the contribution from both the CISS molecule and the ferromagnet, and obtain

\[
T_{12}(\Rightarrow) = T_{CISS} \cdot \left( I - R_{FM}(\Rightarrow) \cdot R_{CISS} \right)^{-1} \cdot T_{FM}.
\]

The transmission probability between the two contacts is therefore

\[
T_{12}(\Rightarrow) = \begin{pmatrix} 1, 1 \end{pmatrix} T_{12}(\Rightarrow) \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

and substituting the matrices gives

\[
T_{12}(\Rightarrow) = \frac{a(1 + P_{FM})}{1 + a + b + P_{FM}(1 - a - b)} + \frac{d(1 - P_{FM})}{1 + c + d - P_{FM}(1 - c - d)}.
\]

Reversing the ferromagnet is equivalent to a sign reversal of \(P_{FM}\), therefore

\[
T_{12}(\Leftarrow) = \frac{a(1 - P_{FM})}{1 + a + b - P_{FM}(1 - a - b)} + \frac{d(1 + P_{FM})}{1 + c + d + P_{FM}(1 - c - d)}.
\]

The broader reciprocity theorem requires\(^{27,28}\)

\[
T_{12}(\Rightarrow) \equiv T_{12}(\Leftarrow).
\]

for all \(0 < |P_{FM}| \leq 1\). This requirement gives

\[a = d, \text{ and } b = c,\]

and therefore,

\[(a + c) - (b + d) = 0,\]
which violates Eqn. S1.

Therefore, we proved that the spin-flip electron reflection process has to exist in order for the CISS effect to exist in the linear transport regime, and this is a direct requirement from the Reciprocity Theorem.

II. FURTHER DISCUSSION ABOUT THE BCB MODEL

A. Derivation of the BCB transmission and reflection matrices

In the BCB model, we consider a CISS molecule as two normal barriers sandwiching an ideal CISS center. The three parts together determine the transmission and reflection matrices of the molecule. We derive these matrices in two steps. First, we calculate the combined contribution of the left-most barrier and the ideal CISS center, as Part 1 (superscript \( P^1 \)). Then, we add the second barrier to Part 1 and calculate the total effect.

We adopt the transmission and reflection matrices for a normal barrier and a CISS center from Fig. 2 in the main text. Here we only discuss P-type CISS molecules, as the AP-type can be derived using the relations given in Fig. 2. For the left-most barrier and the CISS center we have

\[
T_{R}^{P^1} = T_B \cdot (T_{0,R}^P + R_{0,R}^P \cdot R_B \cdot T_{0,R}^P) = \begin{pmatrix} t & 0 \\ t(1-t) & 0 \end{pmatrix},
\]

\[
R_{R}^{P^1} = R_B + T_B \cdot R_{0,R}^P \cdot T_B = \begin{pmatrix} 1-t & 0 \\ t^2 & 1-t \end{pmatrix},
\]

\[
T_{L}^{P^1} = T_{0,L}^P \cdot (T_B + R_B \cdot R_{0,R}^P \cdot T_B) = \begin{pmatrix} 0 & 0 \\ t(1-t) & t \end{pmatrix},
\]

\[
R_{L}^{P^1} = R_{0,L}^P + T_{0,L}^P \cdot R_B \cdot R_{0,R}^P \cdot R_B \cdot T_{0,R}^P = \begin{pmatrix} 0 & 1 \\ (1-t)^2 & 0 \end{pmatrix}.
\]

Note that here we have finite number of reflections between the first barrier and the CISS center because the CISS center is ideal. Now we consider Part 1 as one unit and combine it with the second normal barrier

\[
T_{R}^{P} = T_{R}^{P^1} \cdot (I - R_B \cdot R_{L}^{P^1})^{-1} \cdot T_B = \begin{pmatrix} \sigma/(1-t) & \sigma \\ \sigma & \sigma(1-t) \end{pmatrix},
\]
\[
\mathbb{R}_R^P = \mathbb{R}_R^{P_1} + T_R^{P_1} \cdot \mathbb{R}_B \cdot (\mathbb{I} - \mathbb{R}_L^{P_1} \cdot \mathbb{R}_B)^{-1} \cdot T_L^{P_1} = \begin{pmatrix}
\sigma(1-t)^2 - t + 1 & \sigma(1-t) \\
\sigma/(1-t) & \sigma(1-t)^2 - t + 1
\end{pmatrix},
\]

\[
T_L^P = T_B \cdot (\mathbb{I} - \mathbb{R}_L^{P_1} \cdot \mathbb{R}_B)^{-1} \cdot T_L^{P_1} = \begin{pmatrix}
\sigma(1-t) & \sigma \\
\sigma & \sigma/(1-t)
\end{pmatrix},
\]

\[
\mathbb{R}_L^P = \mathbb{R}_B + T_B \cdot \mathbb{R}_L^{P_1} \cdot (\mathbb{I} - \mathbb{R}_B \cdot \mathbb{R}_L^{P_1})^{-1} \cdot T_B = \begin{pmatrix}
\sigma(1-t)^2 - t + 1 & \sigma/(1-t) \\
\sigma(1-t) & \sigma(1-t)^2 - t + 1
\end{pmatrix},
\]

where \( \sigma = \frac{t^2(1-t)}{-t^4 + 4t^3 - 6t^2 + 4t}. \)

These results show that in a BCB model one parameter \( t \) \((0 < t \leq 1)\) determines the entire set of transmission and reflection probability matrices. Therefore it is possible to plot various transmission or reflection analysis results as functions of \( t \), as shown in the final part of this supplementary information. With different \( t \) values the BCB model is able to represent a large spectrum of CISS molecules with different "strengths" of the CISS effect. However, the matrices are highly symmetric with a single parameter \( t \), indicating that it is still a simplified picture. We will introduce a more generalized model later in this supplementary information.

**B. Discussion on the FM-BCB geometry**

In the main text we discussed that the two-terminal transmission remains constant under magnetization reversal in the FM-BCB geometry, here we illustrate the same result under current reversal.

The transmission and reflection matrices from contact 2 to contact 1 are

\[
T_{21}^{FM-BCB} (\Rightarrow) = T_L^P \cdot \left( \mathbb{I} - \mathbb{R}_F^M (\Rightarrow) \cdot \mathbb{R}_R^{P} \right)^{-1} \cdot T_F^M (\Rightarrow),
\]

\[
\mathbb{R}_{22}^{FM-BCB} (\Rightarrow) = \mathbb{R}_L^P + T_L^P \cdot \mathbb{R}_F^M (\Rightarrow) \cdot \left( \mathbb{I} - \mathbb{R}_R^{P} \cdot \mathbb{R}_F^M (\Rightarrow) \right)^{-1} \cdot T_R^P,
\]

and the corresponding transmission and reflection probabilities are

\[
T_{21}^{FM-BCB} (\Rightarrow) = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} T_{21}^{FM-BCB} (\Rightarrow) \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]
FIG. S1: Normalized total transmission and reflection of a FM-BCB segment as a function of $t$ (BCB barrier transmission). Red and magenta labels (lower curve) are for total transmissions accounting for both spin species for different magnetization orientations and different current flow directions. Blue and cyan labels (upper curve) are for total reflections. The polarization of the ferromagnet is chosen as $P_{FM} = 0.3$, comparable to experimental conditions with Co contacts.

$$
R^{FM-BCB}_{22} (\Rightarrow) = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} R^{FM-BCB}_{22} (\Rightarrow) \begin{pmatrix} 1 \\ 1 \end{pmatrix}.
$$

With these expressions, we can calculate the transmission and reflection probabilities as a function of $t$ (BCB transmission probability) for four situations: two current directions and two magnetization directions, and the result is plotted in Fig. S1. Note that for all four situations, the transmission curves (or the reflection curves) completely overlap with each other, this means that neither magnetization reversal nor current reversal can lead to a signal change in the two-terminal conductance. Furthermore, we can quantitatively analyze the contribution of each spin component in each of the four situations, as shown in the additional figures of this supplementary information.
FIG. S2: Normalized total transmission and reflection of a spin-valve segment as a function of $t$ (BCB barrier transmission). Red and magenta labels (lower curve) are for total transmissions accounting for both spin species for different ferromagnet configurations and different current flow directions. Blue and cyan labels (upper curve) are for total reflections. The polarization of the ferromagnet is chosen as $P_{FM} = 0.3$, comparable to experimental conditions with Co contacts.

C. Discussion on the spin valve geometry

Similar to the FM-BCB geometry, in order to calculate the two-terminal transmission and reflection probability of the spin valve geometry, we first calculate the transmission and reflection matrices for this geometry. For this, we treat the spin valve geometry as a FM-BCB module and a ferromagnet connected in series, and derive

$$T_{12}^{SV} (\Rightarrow, \Rightarrow) = T_{12}^{FM-BCB} (\Rightarrow) \cdot \left( I - R_{FM} (\Rightarrow) \cdot R_{22}^{FM-BCB} (\Rightarrow) \right)^{-1} \cdot T_{FM} (\Rightarrow),$$

$$R_{11}^{SV} (\Rightarrow, \Rightarrow) = R_{11}^{FM-BCB} (\Rightarrow) + T_{12}^{FM-BCB} (\Rightarrow) \cdot R_{FM} (\Rightarrow) \cdot \left( I - R_{22}^{FM-BCB} (\Rightarrow) \cdot R_{FM} (\Rightarrow) \right)^{-1} \cdot T_{21}^{FM-BCB} (\Rightarrow),$$
\[
T_{21}^{SV}(\Rightarrow, \Rightarrow) = T_{FM}(\Rightarrow) \cdot \left( I - R_{22}^{FM-BCB}(\Rightarrow) \cdot R_{FM}(\Rightarrow) \right)^{-1} \cdot T_{21}^{FM-BCB}(\Rightarrow),
\]

\[
R_{22}^{SV}(\Rightarrow, \Rightarrow) = R_{FM}(\Rightarrow) + T_{FM}(\Rightarrow) \cdot R_{22}^{FM-BCB}(\Rightarrow) \cdot \left( I - R_{FM}(\Rightarrow) \cdot R_{22}^{FM-BCB}(\Rightarrow) \right)^{-1} \cdot T_{FM}(\Rightarrow),
\]

where the two arrows in the brackets on the left-hand side of the equations indicate the magnetization direction of the two ferromagnets respectively. For the case where one of the ferromagnetic barriers is reversed, we can substitute the corresponding magnetization direction with an opposite arrow.

For the two-terminal transmission and reflection probabilities of this geometry, we have

\[
T_{ij}^{SV} = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} \cdot T_{ij}^{SV} \cdot \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

\[
R_{ij}^{SV} = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} \cdot R_{ij}^{SV} \cdot \begin{pmatrix} 1 \\ 1 \end{pmatrix},
\]

and we can calculate these probabilities as a function of \( t \) (BCB transmission probability) for eight situations: the two ferromagnets each with two magnetization directions, and two opposite current directions, as shown in Fig. S2. Note that for all eight situations, the transmission curves (or the reflection curves) completely overlap with each other, this means that neither magnetization reversal (for either ferromagnet) nor current reversal can lead to a signal change in the two-terminal conductance. Furthermore, we can quantitatively analyze the contribution of each spin component in this geometry, as shown in the additional figures of this supplementary information.

**D. Discussion on injection and detection coefficients**

In the main text we derived the injection and detection coefficients \( k_{inj} \) and \( k_{det} \) for the four-terminal geometry shown in Fig. 6 textbfa), and we showed that the product of these two geometries represents the ratio between four-terminal and two-terminal resistances. The injection coefficient depends both
FIG. S3: Injection coefficient $k_{\text{inj}}$ as a function of $t$ (BCB barrier transmission) for various $t_B$ (contact barrier transmission), for the geometry described in Fig. 6a) in the main text.

FIG. S4: Detection coefficient $k_{\text{det}}$ as a function of $t$ (BCB barrier transmission) for the geometry described in Fig. 6a) in the main text.

on the BCB barrier transmission $t$ and the transmission probability $t_B$ of the barrier at contact 2, and the detection coefficient only depends on the BCB barrier transmission $t$. This
FIG. S5: The ratio between four-terminal and two-terminal resistances as a function of $t$ (BCB barrier transmission) for various $t_B$ (contact barrier transmission), for the geometry described in Fig. 6a) in the main text.

The difference is due to our different assumptions about the injection and detection contacts. Here we plot the injection coefficient $k_{inj}$, the detection coefficient $k_{det}$, and the 4T/2T resistance ratio $R_{4T}/R_{2T}$ as a function of $t$ (transmission probability of the barrier in the BCB molecule). Especially, for $k_{inj}$ and $R_{4T}/R_{2T}$, we set $t_B$ (transmission probability of the barrier in contact 2) to a few different values and illustrate its influence.

E. Discussion on spin-charge conversion

In the main text we illustrated the spin-charge conversion property of CISS molecules. The chemical potential vectors in the two nodes in Fig. 7 of the main text are related to each other following

$$\mu_B = \mu_A T_R^F (\mathbb{I} - \mathbb{R}_L^F)^{-1},$$

which is Eqn. 16 in the main text. We also introduced the scalers of charge chemical potential $\mu_n$ (average of two spins) and spin accumulation $\mu_s$ (difference of two spins). Here we show how these scaler chemical potentials relate to each other.

A vector chemical potential of a node (same for both nodes in this geometry) can be
rewritten in the following fashion

\[ \mu = \left( \mu_{\rightarrow}, \mu_{\leftarrow} \right) = \left( \mu_n + \frac{1}{2} \mu_s, \mu_n - \frac{1}{2} \mu_s \right) = \mu_n \left( 1, 1 \right) + \frac{1}{2} \mu_s \left( 1, -1 \right) \]

Substituting the above type of expression for both \( \mu_A \) and \( \mu_B \), and apply to Eqn. S2 we obtain

\[ \Delta \mu_n \left( 1, 1 \right) + \frac{1}{2} \Delta \mu_s \left( 1, -1 \right) = \mu_A \left( I - T^P_R (I - R^P_L)^{-1} \right), \tag{S3} \]

where \( \Delta \mu_n = \mu_{nA} - \mu_n \) and \( \Delta \mu_s = \mu_{sA} - \mu_{sB} \).

For the BCB model,

\[ I - T^P_R (I - R^P_L)^{-1} = \begin{pmatrix} 1 + \frac{1}{t-2} & \frac{1}{t-2} \\ -1 - \frac{1}{t-2} & -1 \end{pmatrix} \]

where \( t \) is the BCB transmission probability. Substituting this matrix into Eqn. S3 gives

\[ \Delta \mu_n \left( 1, 1 \right) + \frac{1}{2} \Delta \mu_s \left( 1, -1 \right) = \left( (1 + \frac{1}{t-2}) \mu_{sA}, \frac{1}{t-2} \mu_{sA} \right). \]

Note that the charge chemical potentials in node \( A \) \( (\mu_{nA}) \) drops out of the equation. This gives rise to later Eqn. S4a. We emphasize that this is a unique result for the BCB model thanks to the fact that the BCB model is a simplified picture and the matrices are highly symmetric.

The above vector equation is equivalent to two equations

\[ \Delta \mu_n + \frac{1}{2} \Delta \mu_s = (1 + \frac{1}{t-2}) \mu_{sA}, \]

\[ \Delta \mu_n - \frac{1}{2} \Delta \mu_s = \frac{1}{t-2} \mu_{sA}. \]

From these equations we can derive

\[ \Delta \mu_s = \mu_{sA}, \tag{S4a} \]

\[ \Delta \mu_n = -k_{\text{conv}} \Delta \mu_s, \tag{S4b} \]

where \( k_{\text{conv}} = t/(4 - 2t) \) \( (0 < k_{\text{conv}} \leq 0.5) \) is the spin-charge conversion coefficient.

Eqn. S4a shows that \( \mu_{sB} = \mu_{sA} - \Delta \mu_s = 0 \), meaning that a spin accumulation in node \( A \) cannot generate a spin accumulation in node \( B \). This result is special for the BCB
model and does not hold for a more generalized CISS model (as will be introduced later in this supplementary information). Eqn. S4b shows that the charge voltage across a BCB molecule linearly depends on the spin accumulation difference across the molecule, and vice versa (spin-charge conversion). The conversion coefficient \( k_{\text{conv}} \) depends on \( t \) (BCB barrier transmission). Notably, for BCB molecules this coefficient has the same value as the detection coefficient \( k_{\text{conv}} = k_{\text{det}} \). This is because the result \( \mu_{sB} = 0 \) makes node \( B \) equivalent to a contact: it is not able to distinguish between two spin components. For a more generalized model (as will be introduced later in this supplementary information) the two coefficient take different values. These conclusions were mentioned in the main text, here we showed the mathematical proof.

F. Discussion on nonlocal signals

We first discuss the non-local resistance measured with device geometries shown in Fig. 8a) in the main text. In this picture the axis of the CISS molecules are vertical rather than horizontal, therefore the spin orientations are described as spin-up or spin-down. We define spin-up as the majority, as with spin-right in other discussions. As a result, the spin accumulation is defined as \( \mu_s = (\mu_\uparrow - \mu_\downarrow) \).

For the case where no magnetic materials are used, we note the injected spin accumulation underneath contact 1 as \( \mu_{s,\text{inj}} \)

\[
\mu_{s,\text{inj}} = -k_{\text{inj}}\mu_1 = -ek_{\text{inj}}I_{\text{inj}}R_{\text{inj}};
\]

where the minus sign is due to the fact that the current is going downwards through CISS molecules into graphene, as a result the injected spin accumulation is negative (mostly spin-down). The resistance

\[
R_{\text{inj}} = R_{12} = \frac{\mu_1 - \mu_2}{eI_{\text{inj}}}
\]

is the resistance measured between the two injection contacts 1 and 2.

Inside graphene, the spin accumulation diffuses to all directions, the spin accumulation at the detection contact is thus

\[
\mu_{s,\text{det}} = \frac{1}{2}\mu_{s,\text{inj}}e^{-\frac{d}{\lambda_s}},
\]

where \( \lambda_s \) is the spin diffusion length in graphene, and \( d \) is the distance between the inner injection and detection contacts (1 and 4), and we have assumed that this distance is much
larger than the separation of the two injection contacts or the separation of the two detection contacts. The factor 1/2 is because the detection contact is only on one side of the injection contact, while the diffusion happens symmetrically to both sides.

Further, the voltage detected by the detection contacts is (following Eqn. 13 of the main text)

\[ V_{\text{det}} = \frac{1}{e} k_{\text{det}} \mu_{s, \text{det}}. \]

With this, we have

\[ R_{nl} = \frac{V_{\text{det}}}{I_{\text{inj}}} = -\frac{1}{2} k_{\text{inj}} k_{\text{det}} R_{\text{inj}} e^{-\frac{d}{\lambda_s}}, \tag{S5} \]

as in Eqn. 17 of the main text.

For the case where the spin injection is obtained through a ferromagnet, the spin injection becomes

\[ \mu_{s, \text{inj}} = \pm e P_{FM} I_{\text{inj}} R_{\lambda}, \]

where \( P_{FM} \) is the polarization of the ferromagnet (with magnetization direction out-of-plane), and \( R_{\lambda} \) is the spin resistance of graphene. This spin resistance is determined by the spin relaxation length in graphene and the shape of the graphene channel, and is defined as

\[ R_{\lambda} = \frac{R_{sq} \lambda_s}{W}, \]

where \( R_{sq} \) is the square resistance of graphene and \( W \) is the width of the graphene channel (assuming the channel width remains the same across the spin diffusion length). The sign of the injected spin accumulation is determined by the magnetization direction of the ferromagnet, with magnetization-up for positive spin accumulation and magnetization-down for negative spin accumulation.

The diffusion and detection mechanisms are the same as in the previous case. Therefore, the non-local resistance for this situation is

\[ R_{nl} = \pm \frac{1}{2} k_{\text{det}} P_{FM} R_{\lambda} e^{-\frac{d}{\lambda_s}}, \tag{S6} \]

as in Eqn. 18 in the main text. With the help of the ferromagnet it is possible to switch the sign of the non-local resistance.

Next, for the device shown in Fig. 8b) in the main text, the CISS molecules are aligned in-plane of the device, therefore we assume the ferromagnet also has in-plane magnetization, and we describe spin accumulation again as \( \mu_s = \mu_\rightarrow - \mu_\leftarrow \). We simplify the discussion by
assuming the spin injection is mainly contributed by the ferromagnet, and the spin detection
is achieved through the spin-charge conversion mechanism of the CISS molecules. The spin
accumulation underneath the detection contacts can be generated by two mechanisms: the
spin diffusion in graphene (as in the previous case), and the spin-charge conversion between
the injection node and the detection node. However, under the BCB model, the spin-charge
conversion does not contribute to a spin accumulation underneath the detector contacts.
Therefore, we only consider the spin diffusion mechanism. Similar to the previous case, we
can derive the nonlocal signal

\[ R_{nl} = \pm \frac{1}{2} k_{\text{conv}} P_{FM} R_{\lambda} e^{-\frac{d}{\lambda_s}}, \]  

(S7)

where \( k_{\text{conv}} \) is the spin-charge conversion coefficient described before, but here it concerns
the proximity-induced CISS effect in the graphene channel, rather than the CISS molecules
themselves. Due to this proximity effect, the diffusion length \( \lambda_s \) and the spin resistance \( R_\lambda \)
of graphene may differ from the previous case.

III. GENERALIZED CISS MODEL.

The BCB model is a simplified model where the spin-dependent characteristics of a CISS
molecule are exclusively originated from an ideal CISS center. However, the assumption of
having an ideal spin-flip core in a molecule is not accurate. Therefore, we assume a general
form of transmission and reflection matrices,

\[ T_R^P = \begin{pmatrix} a & b \\ c & d \end{pmatrix}, \quad R_R^P = \begin{pmatrix} A & B \\ C & D \end{pmatrix}, \]

where

\[ 0 \leq a, b, c, d, A, B, C, D \leq 1, \]  

(S8a)

\[ a + b + A + B = c + d + C + D = 1, \]  

(S8b)

\[ a + c > b + d. \]  

(S8c)

Here the first restriction Eqn. S8a comes from the fact that all matrix elements are proba-
bilities. The second restriction Eqn. S8b addresses that for each spin component, the sum
of its probabilities of being transmitted and being reflected equals 1. The third restriction
Eqn. S8c is the conceptual description of the CISS effect, which shows that a spin polarization arises after transmission through a CISS molecule. This restriction is similar to Eqn. S1, but here we determine a sign of the polarization because we have assumed the chirality ($P$-type) of the molecule and the electron flow direction ($R$ for rightwards).

We still assume that the molecule has $C_2$ symmetry, so that the transmission and reflection matrices for reversed current can be written as

$$T^P_L = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \cdot T^P_R \cdot \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix},$$

$$R^P_L = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \cdot R^P_R \cdot \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}.$$ 

The matrices for $AP$-type molecules can be derived according to Fig. 2 in the main text. Before we proceed with mathematical proof, we discuss the validity of these symmetry assumptions.

Since we only consider the electron transport in CISS molecules, the $C_2$ symmetry that we require here is only referring to the symmetry in electron transport. Note that a CISS molecule does not have to have a structural two-fold rotational symmetry in order to have the symmetry for electron transport. Furthermore, we discuss only the linear regime, which by definition requires such symmetry. As for highly asymmetrical molecules, we can consider it as a combination of a symmetrical part which contributes to CISS, and a normal barrier.

As for the symmetry relations between the two chiralities, it is by definition that the two chiral enantiomers are exact mirror images of each other, and thereby select opposite spins with equal probability.

We apply the reciprocity theorem to a geometry similar to Fig. 4 in the main text, but now the BCB molecule is replaced by a generalized CISS molecule. The general reciprocity theorem requires that the two-terminal resistance remains unchanged under magnetization reversal regardless of the polarization of the ferromagnet. Therefore, we assume the ferromagnet is 100% polarized for convenience. Following the same steps as in the FM-BCB geometry, the two terminal transmission probabilities considering two magnetization directions and two current directions become

$$T_{12}(\Rightarrow) = a + b + (c + d) \frac{B}{1 - D},$$
where the arrows indicate the magnetization direction of the ferromagnet, and the subscripts indicate the electron flow direction. The reciprocity theorem requires these four expressions to have the same value

\[ T_{12}(\rightarrow) = T_{12}(\leftarrow) = T_{21}(\rightarrow) = T_{21}(\leftarrow) = T, \]

which can be written in a matrix form

\[
\begin{pmatrix}
1 & 1 & \frac{B}{1-D} & \frac{B}{1-D} \\
\frac{C}{1-A} & \frac{C}{1-D} & 1 & 1 \\
\frac{C}{1-A} & 1 & \frac{C}{1-D} & 1 \\
1 & \frac{B}{1-A} & 1 & \frac{B}{1-A}
\end{pmatrix}
\begin{pmatrix}
a \\
b \\
c \\
d
\end{pmatrix}
= T
\begin{pmatrix}
1 \\
1 \\
1 \\
1
\end{pmatrix}.
\]

In order for this equation to be self-consistent, relation

\[ A = D \]

is required. Consequently, we can derive

\[ b = c, \]

\[ a - d = C - B. \]

Therefore, the generalized form of the transmission and reflection matrices of a CISS molecule is

\[ T_R^P = \begin{pmatrix}
a & b \\
b & a(1-s)
\end{pmatrix}, \quad R_R^P = \begin{pmatrix}
A & B \\
B + as & A
\end{pmatrix}, \quad (S9)\]

with \( a + b + A + B = 1 \), and \( s \) (\( 0 \leq s \leq 1 \)) being a quantitative description of the strength of the CISS effect in a molecule.

Next, we discuss the results given by this generalized model for the geometries discussed in the main text, and compare it with the BCB model. We arbitrarily choose the following transmission and reflection matrices for the generalized model

\[ T_R^P = \begin{pmatrix}
0.6 & 0.15 \\
0.15 & 0.6(1-s)
\end{pmatrix}, \quad R_R^P = \begin{pmatrix}
0.2 & 0.05 \\
0.05 + 0.6s & 0.2
\end{pmatrix}, \quad \text{with } s = 0.5. \]
FIG. S6: Normalized total transmission and reflection of a FM-CISS segment as a function of $s$ (generalized CISS strength). Red and magenta labels (lower curve) are for total transmissions accounting for both spin species for different magnetization orientations and different current flow directions. Blue and cyan labels (upper curve) are for total reflections. The polarization of the ferromagnet is chosen as $P_{FM} = 0.3$, comparable to experimental conditions with Co contacts.

and use parameter $s$ as an variable to tune the CISS strength.

For the FM-BCB geometry (which now becomes FM-CISS geometry), we plot the two-terminal transmission and reflection probabilities as a function of $s$ for four situations: two magnetization directions and two current directions. The results show that all four situations give identical results for all CISS strength $s$, as shown in Fig. S6. These results are the same as with the BCB model, and are consistent with the reciprocity theorem.

For the spin valve geometry, we plot the two-terminal transmission and reflection probabilities as a function of $s$ for eight situations: two ferromagnets each with two magnetization directions, and two current directions. The results are shown in Fig. S7. Unlike the BCB model, here we find different transmission and reflection probabilities for cases where the two magnetization directions are parallel vs. anti-parallel. The transmission probability is in general higher when the two ferromagnets are magnetized parallel compared to anti-parallel. Notably, the difference between the two configurations decreases as the CISS
FIG. S7: Normalized total transmission and reflection of a spin-valve segment as a function of $s$ (generalized CISS strength). Red and magenta labels (lower two curves) are for total transmissions accounting for both spin species for different ferromagnet configurations and different current flow directions. Blue and cyan labels (upper two curves) are for total reflections. The polarization of the ferromagnet is chosen as $P_{FM} = 0.3$, comparable to experimental conditions with Co contacts.

strength $s$ increases. This trend is because of an increasing chance of electrons encountering spin-flip reflections when $s$ increases. This also explains why in a BCB model, where an ideal spin-flip process is present, switching the magnetization directions does not lead to any conductance variation. The direction of current has no effect on the transmission and reflection probabilities, as required by the linear regime.

In terms of four terminal geometries, we calculate here the spin injection and detection coefficients using the formulas derived in the main text, and we show these coefficient as a function of the CISS strength $s$.

Fig. S8 shows the injection coefficient as a function of $s$ for a few $t_B$ values. We point out that $k_{inj}$ does not always linearly depend on $s$, although in this figure it seems to be the case. The two curves for $t_B = 0.7$ and $t_B = 0.9$ are not distinguishable due to a high degree of overlap.

Fig. S9 shows the detection coefficient as a function of $s$, and Fig. S10 shows the ratio
FIG. S8: Injection coefficient $k_{inj}$ as a function of $s$ (generalized CISS strength) for various $t_B$ (contact barrier transmissiiion), for the geometry described in Fig. 6a) in the main text.

FIG. S9: Detection coefficient $k_{det}$ as a function of $s$ (generalized CISS strength) for the geometry described in Fig. 6a) in the main text.

between four-terminal and two-terminal resistances as a function of $s$. The behaviors in both plots resemble those of the BCB model, but are quantitatively different.
FIG. S10: The ratio between four-terminal and two-terminal resistances as a function of $s$ (generalized CISS strength) for various $t_B$ (contact barrier transmission), for the geometry described by Fig. 6a) in the main text.

Last but not the least, we discuss the spin-charge conversion property of the generalized CISS molecule. Substituting the corresponding transmission and reflection matrices into Eqn. [S3] and solving the vector equation, one can obtain relations among $\Delta \mu_n$, $\Delta \mu_s$ and $\mu_A$. Unlike the BCB model, here it is not possible to drop out either $\mu_{nA}$ or $\mu_{sA}$ from the equation. The final result gives

$$k_{conv} = -\frac{\Delta \mu_n}{\Delta \mu_s} = \frac{(A - B - 1)s\mu_{nA} + \frac{1}{2}(b + B)s\mu_{sA}}{2(A + B - 1)s\mu_{nA} + (b + B)(-2 + 2A + 2B + s)\mu_{sA}},$$

where $a, b, A, B, s$ are the parameters in the transmission and reflection matrices of the generalized CISS model. This equation shows that the a non-zero charge voltage difference can give rise to a spin accumulation difference across a generalized CISS molecule, and *vice versa* (spin-charge conversion). Interestingly, the conversion coefficient $k_{conv}$ depends not only on the transmission and reflection matrices of the molecule, but also on the spin accumulation in the nodes connected to the molecule. If $s = 0$, i.e. the molecule does not exhibit any CISS effect, the spin-charge conversion property also diminishes ($k_{conv} = 0$).

With these discussions, we demonstrated that a generalized CISS molecule shows differences from the simplified BCB model. Nevertheless, these differences are rather quantitative
than qualitative, and are not easily measurable in experiments. Furthermore, the general-
ized model also introduces extra degrees-of-freedom to calculations as it uses four variables
to describe a CISS molecule, compared to one in the BCB model. Having taken the above
into consideration, in the main text we only showed results of the BCB model.
IV. ADDITIONAL FIGURES

In this section we show additional figures for the BCB model.

Fig. S11 to Fig. S14 show the transmission and reflection probability of each spin component for each of the four situations (two magnetization directions and two current directions) of the FM-BCB geometry. In these four situations, even though the total transmissions (or reflections) concerning both spin components are identical, the transmissions and reflections for each spin component, and hence the spin polarizations of the electrons entering the contact are different. This difference cannot be measured because the contacts cannot distinguish between spin components.

Fig. S15 to Fig. S22 show the same analysis for the eight situations (two ferromagnets each with two magnetization directions, and two current directions) of the spin valve geometry. Similarly, spin polarization can be generated at the contacts, and are different for all situations. However, due to the limitation of the two-terminal geometry, the total transmissions (or reflections) concerning both spin components remain the same for all eight situations.

FIG. S11: Normalized one-spin transmission or reflection for the FM-BCB case, where the magnetization of the ferromagnet is "u" (pointing to the right) and electron flow "forward" (from left to right).
FIG. S12: Normalized one-spin transmission or reflection for the FM-BCB case, where the magnetization of the ferromagnet is "u" (pointing to the right) and electron flow "reverse" (from right to left).

FIG. S13: Normalized one-spin transmission or reflection for the FM-BCB case, where the magnetization of the ferromagnet is "d" (pointing to the left) and electron flow "forward" (from left to right).
FIG. S14: Normalized one-spin transmission or reflection for the FM-BCB case, where the magnetization of the ferromagnet is "d" (pointing to the left) and electron flow "reverse" (from right to left).

FIG. S15: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "uu" (both pointing to the right) and electron flow "forward" (from left to right).
FIG. S16: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "uu" (both pointing to the right) and electron flow "reverse" (from right to left).

FIG. S17: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "ud" (the first one pointing to the right, and the second one to the left) and electron flow "forward" (from left to right).
FIG. S18: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "ud" (the first one pointing to the right, and the second one to the left) and electron flow "reverse" (from right to left).

FIG. S19: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "du" (the first one pointing to the left, and the second one to the right) and electron flow "forward" (from left to right).
FIG. S20: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "du" (the first one pointing to the left, and the second one to the right) and electron flow "reverse" (from right to left).

FIG. S21: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "dd" (both pointing to the left) and electron flow "forward" (from left to right).
FIG. S22: Normalized one-spin transmission or reflection for the spin valve case, where the magnetization of the ferromagnet is "dd" (both pointing to the left) and electron flow "reverse" (from right to left).