Injection and detection of spin in a semiconductor by tunneling via interface states

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Injection and detection of spin accumulation in a semiconductor having localized states at the interface is evaluated. Spin transport from a ferromagnetic contact by sequential, two-step tunneling via interface states is treated not in itself, but in parallel with direct tunneling. The spin accumulation $\Delta\mu^{ch}$ induced in the semiconductor channel is not suppressed, as previously argued, but genuinely enhanced by the additional spin current via interface states. Spin detection with a ferromagnetic contact yields a weighted average of $\Delta\mu^{ch}$ and the spin accumulation $\Delta\mu^{ls}$ in the localized states. In the regime where $\Delta\mu^{ls}/\Delta\mu^{ch}$ is largest, the detected spin signal is insensitive to $\Delta\mu^{ls}$ and the ferromagnet probes the spin accumulation in the semiconductor channel.

Spin polarization can be created in non-magnetic semiconductors by spin-polarized tunneling from a ferromagnetic contact. This powerful, robust and technologically viable approach has been demonstrated in various semiconductors, including silicon and germanium and at room temperature\textsuperscript{2–11}. Considerable discussion has arisen because the magnitude of the spin accumulation induced in the semiconductor is consistently in disagreement with the theory for spin injection and spin diffusion\textsuperscript{12–15}. The detected spin signal is often found to be orders of magnitude larger than expected, particularly for three-terminal devices in which the spin signal is induced and probed by a single magnetic tunnel contact\textsuperscript{15,6,9–11,16,17}. But in some Si and Ge based devices with non-local geometry (with separate spin injection and detection contacts) the spin signal is significantly smaller than predicted if reasonable values of the contact tunnel spin polarization are used\textsuperscript{18}. Understanding the origin of these puzzling results is indispensable because spin injection and detection by a magnetic tunnel contact is a cornerstone of semiconductor spintronics.

While there are indications that the standard theory for spin injection does not capture all the physics\textsuperscript{2,3,4,8,12–15}, and lateral inhomogeneity of the tunnel current may also contribute\textsuperscript{16}, it is also heavily debated whether localized states near the semiconductor interface play a role. These can give rise to resonant tunneling, non-resonant scattering and inelastic tunneling and thereby reduce or even invert the tunneling spin polarization\textsuperscript{18–28}. In a different vein, ferromagnet/insulator/semiconductor structures under photo-excitation were described by sequential, two-step transport via interface states with their own spin accumulation and spin relaxation rate\textsuperscript{29,30}. The states are separated from the ferromagnet by a tunnel barrier and from the semiconductor bulk by a Schottky barrier and for the latter, transport by thermionic emission was considered. Just as for spin injection into non-degenerate semiconductors\textsuperscript{31}, this severely compromises the spin selectivity of the contacts. Recently, Tran et al. also considered spin injection by two-step, sequential transport, but assumed tunneling across the barrier between localized states and semiconductor\textsuperscript{32}. Importantly, it was predicted that the spin accumulation $\Delta\mu^{ls}$ in the localized states can be much larger than the spin accumulation $\Delta\mu^{ch}$ induced in the semiconductor channel, albeit under certain conditions.

If two-step tunneling via interface states indeed plays a role, it may have crucial implications for the injection and detection of spin in a multitude of devices that employ tunnel contacts. Two pertinent questions are: (i) what is the effect of two-step tunneling via interface states on the spin accumulation that is created in the semiconductor? (ii) how does two-step tunneling affect the detection of spin accumulation in the semiconductor by a magnetic contact? Tran et al. predict that the spin accumulation in the semiconductor can be severely suppressed if spins relax in the intermediate localized states\textsuperscript{32}. They also predict that a ferromagnetic contact does not probe $\Delta\mu^{ch}$, but instead $\Delta\mu^{ls}$, which can be much larger than $\Delta\mu^{ch}$, particularly for small density of localized states. Given the implications, it is unfortunate that it has become practice to automatically attribute enhanced spin signals seen in experiment to spin accumulation in interface states, without examining whether the conditions to produce an enhancement are fulfilled, and without critical tests, for instance, varying specific parameters and observing whether the experimental data follows the expected trends.

To address the effect of interface states, a correct prediction of their impact on spin transport is required. It is shown here that Tran’s model\textsuperscript{32} and the trends it predicts need significant revision, because a basic assumption, namely that all the tunnel current between ferromagnet and semiconductor is through localized states, is not generally valid. Here we treat two-step tunneling via interface states in parallel with direct tunneling. We show that the spin accumulation in the semiconductor channel is not suppressed, but genuinely enhanced by the additional spin current via interface states. We also find that spin detection with a ferromagnetic contact yields a weighted average of $\Delta\mu^{ch}$ and $\Delta\mu^{ls}$, which shifts depending on the ratio of direct and two-step tunneling current. Spin accumulation in interface states only enhances the detected spin signal in the intermediate regime where both current components are comparable, and only if the localized states are separated from the semiconductor by a barrier with sufficiently large resistance.
FIG. 1: Energy band diagram of a ferromagnet/insulator/semiconductor junction with localized states (LS) separated from the ferromagnet (FM) by tunnel barrier 1, and from the semiconductor (SC) by tunnel barrier 2. A spin accumulation exists in the semiconductor channel ($\Delta \mu_{ch}$) and in the localized states ($\Delta \mu_{ls}$). The circuit depicts the spin currents and resistances, with $\bullet$ representing spin sinks due to spin relaxation in LS and SC. The FM is the spin source.

The system has three sections, a ferromagnet, localized states with spin-integrated density $D_{ls}$ (in states eV$^{-1}$m$^{-2}$), and a semiconductor channel (Fig. 1). The potential of the ferromagnet is taken to be the zero. The spin-averaged potentials of the semiconductor channel and the localized states are $V$ and $V_{ls}$, respectively. For direct tunneling, the charge current $I$ and spin current $I_s$ are (see also appendix):

$$I = GV - PG_1G\left(\frac{\Delta \mu_{ch}}{2}\right)$$

$$I_s = PG_1GV - G\left(\frac{\Delta \mu_{ch}}{2}\right)$$

where $G$ is the total (spin-averaged) tunnel conductance and $P_G$ is the spin polarization of the conductance. Note that the spin accumulation decays into the semiconductor and that $\Delta \mu_{ch}$ is the value at the interface, since this determines the tunneling process. For two-step tunneling via localized states, we denote the charge and spin current between ferromagnet and localized states by $I_1$ and $I_{s,1}$, respectively, the total conductance by $G_1$ and the conductance spin polarization by $P_{G1}$. For the second tunnel step between localized states and semiconductor channel, the charge and spin current are denoted by $I_2$ and $I_{s,2}$, the total tunnel conductance is $G_2$, and the conductance is unpolarized since neither localized states nor semiconductor is ferromagnetic. The charge and spin currents for two-step tunneling are (see also the appendix):

$$I_1 = G_1 V^{ls} - P_{G1}G_1\left(\frac{\Delta \mu_{ls}}{2}\right)$$

$$I_{s,1} = P_{G1}G_1V^{ls} - G_1\left(\frac{\Delta \mu_{ls}}{2}\right)$$

$$I_2 = G_2(V - V^{ls})$$

$$I_{s,2} = G_2\left(\Delta \mu_{ls} - \Delta \mu_{ch}\right)$$

Since direct and two-step tunneling occur in parallel, $\Delta \mu_{ch}$ is determined by the total spin current $I_s + I_{s,2}$ into the channel, where $I_{s,2}$ is proportional to the difference between $\Delta \mu_{ls}$ and $\Delta \mu_{ch}$. The spin accumulation in the localized states gives rise to spin relaxation and an associated spin current $I_{ls}^s = e(N_{ls}^+ - N_{ls}^-)/\tau_{ls}^s$, where $N_{ls}^\sigma$ is the number of electrons with spin $\sigma$ in the localized states, and $\tau_{ls}^s$ is the spin-relaxation time in the localized states. Note that $I_{ls}^s$ is defined in units of electron angular momentum $\hbar/2$ transferred per unit time, instead of spin flips per unit time. The spin resistance of the localized states is $r_{ls}^s = \tau_{ls}^s/(eD_{ls})$, such that $\Delta \mu_{ls} = 2I_s^s r_{ls}^s$. Similarly, spin relaxation in the
A semiconductor interface produces a spin-relaxation spin current \( I_{ch} \) that is related to the spin accumulation by the spin resistance \( r_{ch}^s \) of the semiconductor: \( \Delta \mu_{s \rightarrow ch} = 2 I_{ch} r_{ch}^s \). The relations for \( r_{ls}^s \) and \( r_{ch}^s \), together with eqns. (1)-(6), define the system. The three unknown quantities (\( \Delta \mu_{s \rightarrow ch}, \Delta \mu_{s \rightarrow ls} \) and \( V_{ls} \)) are obtained from the following three conditions: (i) \( I_{ls}^s = I_s + I_{ls} \), (ii) \( I_{ls}^c = I_s - I_{ls} \), and (iii) \( I_l = I_2 \). Condition (i) says that in a steady state, the spin relaxation spin current in the semiconductor is equal to the total spin current injected into it (sum of \( I_s \) and \( I_{ls} \)). Condition (ii) states that the spin relaxation spin current in the localized states must be equal to the difference of the spin current \( I_{s,1} \) injected into it from the ferromagnet and the spin current \( I_{s,2} \) that leaks away into the semiconductor. Charge conservation yields condition (iii). The solutions for the spin accumulations are:

\[
\Delta \mu_{ls} = \frac{\beta_{ls} P_{G1} + P_G}{\frac{1}{G} R_{tun} - 1} \left( \frac{2 R_2}{R_{tun} R_1 + R_2} \right) V \tag{7}
\]

\[
\Delta \mu_{ch} = \frac{\beta_{ch} P_{G1} + P_G}{\frac{1}{G} R_{tun} - 1} \left( \frac{2 R_2}{R_1 + R_2} \right) V \tag{8}
\]

where we defined the resistances \( R_{tun} = 1/G, R_1 = 1/G_1, R_2 = 1/G_2 \) and the dimensionless parameters:

\[
\beta_{ch} = \frac{R_{tun} R_2 + r_s^c (R_2 + R_{tun})}{r_s^c (R_1 + R_2)} \approx \frac{R_{tun} (R_2 + r_s^c)}{R_1 r_s^c} \tag{9}
\]

\[
\beta_{ls} = \frac{R_1 R_2 (R_1 + R_2) + r_s^c [(R_1 + R_2)^2 - (P_{G1} R_2)^2]}{r_s^c (R_1 + R_2)} \approx \frac{R_1 (R_2 + r_s^c)}{R_{tun} r_s^c} \tag{10}
\]

The approximate forms of \( \beta_{ch} \) and \( \beta_{ls} \) are obtained when \( R_1 \gg R_2 \), which applies to localized states at or near the semiconductor interface. If \( R_1 \gg R_2 \), eqns. (7) and (8) reduce to:

\[
\Delta \mu_{ls} = \left( \frac{2 r_s^c}{R_1} \right) P_{G1} V + \left( \frac{r_s^c}{R_2 + r_s^c} \right) \left( \frac{2 r_s^c}{R_{tun}} \right) P_G V \tag{11}
\]

\[
\Delta \mu_{ch} = \frac{r_s^c}{R_2 + r_s^c} \left( \frac{2 r_s^c}{R_1} \right) P_{G1} V + \frac{r_s^c}{R_2 + r_s^c} \left( \frac{2 r_s^c}{R_{tun}} \right) P_G V \tag{12}
\]

where \( r_s^{eff} = r_s^c (R_2 + r_s^c)/ (r_s^c + R_2 + r_s^c) \) as in the work of Tran et al.\(^{15} \). It represents the effective spin resistance of the system of localized states and semiconductor channel, coupled by a tunnel resistance \( R_2 \).

The spin accumulations have a contribution from two-step tunneling (proportional to \( P_{G1} \)) and a contribution that arises from direct tunneling (proportional to \( P_G \)). The latter disappears for \( R_{tun} \to \infty \), for which eqns. (11) and (12) reduce to that obtained in Tran’s model.\(^{15} \) In that case one finds that the spin accumulation is governed by \( r_s^{eff} \) instead of \( r_s^c \), and that \( \Delta \mu_{ls}/\Delta \mu_{ch} \) equals \( 1 + R_2/r_s^c \), which can be much larger than unity when \( R_2 > r_s^c \). Moreover, \( \Delta \mu_{ch} \) becomes vanishingly small when \( R_2 > r_s^c \), \( r_s^c \), corresponding to the situation where spins relax in the localized states before escaping into the semiconductor. In Tran’s model, a spin current into the semiconductor is obtained only when spin relaxation in localized states is negligible (\( R_2 < r_s^c \)).

The behavior changes drastically when direct tunneling is included (finite \( R_{tun} \)). The spin current injected into the semiconductor by direct tunneling is approximately \( P_G V/R_{tun} \), and the associated contribution to \( \Delta \mu_{ch} \) (last term in eqn. (12)) exists in addition to the two-step tunneling contribution. In other words, starting with direct tunneling at a given bias voltage \( V \) and then adding localized states, one increases \( \Delta \mu_{ch} \), since extra spin current is injected into the semiconductor by the two-step tunneling. This extra current can also be highly spin polarized (for \( R_2 < r_s^c \)), which is beneficial for creating a large spin accumulation in the semiconductor channel. Even if the spin current from the localized states is negligible (when \( R_2 > r_s^c \), \( r_s^c \)), the spin accumulation induced by direct tunneling still remains.

Our formalism thus demonstrates that neglecting direct tunneling leads to an incorrect prediction of the magnitude of \( \Delta \mu_{ch} \) and to the erroneous conclusion that localized states have a detrimental effect on the spin accumulation in the semiconductor channel. Treating direct and two-step tunneling on an equal footing is thus crucial in order to assess how localized states affect the induced spin polarization.

Next we address how two-step tunneling via interface states affects the detection of a spin accumulation in the semiconductor. Spin detection is typically done by suppressing the spin accumulation via spin precession in a magnetic field perpendicular to the injected spins (Hanle effect). At constant charge current, the resulting change in voltage \( \Delta V_{Hanle} \) across the tunnel contact is, without approximations:

\[
\Delta V_{Hanle} = \frac{R_1 + R_2}{R_1 + R_2 + R_{tun}} \left( \frac{P_G}{2} \right) \Delta \mu_{ch} + \frac{R_{tun}}{R_1 + R_2 + R_{tun}} \left( \frac{P_{G1}}{2} \right) \Delta \mu_{ls} \tag{13}
\]
where $\Delta \mu^{ch}$ and $\Delta \mu^{ls}$ are the values in the absence of a magnetic field (eqns. 7 and 8). The important point is that the Hanle signal is a weighted average of $\Delta \mu^{ch}$ and $\Delta \mu^{ls}$, with a relative contribution determined by the ratio of the resistances associated with direct tunneling ($R_{tun}$) and two-step tunneling ($R_1 + R_2$). When the current is dominated by the localized states ($R_{tun} >> R_1 + R_2$), the first term is zero and the Hanle signal is governed exclusively by $\Delta \mu^{ls}$, as in Tran’s model\textsuperscript{16}. However, when the current due to two-step tunneling is comparable to or smaller than the direct tunneling current, the weight shifts to the term proportional to $\Delta \mu^{ch}$ and any enhancement of the Hanle signal due to localized states disappears. The resistance of the junction is then determined by direct tunneling, and $\Delta V_{Hanle}$ is insensitive to $\Delta \mu^{ls}$ (a large $\Delta \mu^{ls}$ may still exist, but the voltage across the junction does not depend on it). This essential behavior is not captured when one considers only two-step tunneling.

For a given tunnel barrier, the relative weight of direct and two-step tunneling is proportional to the density of localized states because $R_1$ and $R_2$ scale inversely with $D_{ls}$. This can be seen by writing $R_1 = \tau_1^{exc}/(e D_{ls})$ and $R_2 = \tau_2^{exc}/(e D_{ls})$, where $\tau_1^{exc}$ and $\tau_2^{exc}$ are the characteristic time for escape of an electron from, respectively, localized states into the ferromagnet and into the semiconductor channel, as determined by the transmission probability of tunnel barrier 1 and 2. At large $D_{ls}$, the resistance for two-step tunneling is smaller than the resistance for direct tunneling (top panel of Fig. 2). As $D_{ls}$ is reduced, $R_1 + R_2$ increases rapidly and surpasses $R_{tun}$ at a critical value $D_{ls}^{crit}$. Beyond this, direct tunneling dominates. This has a marked effect on the Hanle signal (bottom panel). Tran’s model\textsuperscript{16} predicts increasingly large values of the Hanle signal at smaller $D_{ls}$ (blue curve) because lower $D_{ls}$ means larger spin resistance ($v_{ls} \propto 1/D_{ls}$) and thus a larger spin accumulation in the localized states. However, our full model shows that the Hanle signal goes through a maximum at $D_{ls}^{crit}$, and for smaller $D_{ls}$, the Hanle signal is reduced and approaches the value obtained for pure direct tunneling. We thus find that Tran’s model does not predict the correct variation with $D_{ls}$ and is not valid in the regime where it predicts the largest enhancement of the spin signals - it grossly overestimates the Hanle signal for $D_{ls} < D_{ls}^{crit}$. For large $D_{ls}$, where the signal enhancement is limited, Tran’s model gives approximately the correct value of the Hanle signal, but note that even then it does not predict the correct value of $\Delta \mu^{ch}$, as explained. The value of $D_{ls}^{crit}$ depends on the tunnel probabilities for direct and two-step tunneling through the condition $R_{tun} \approx (\tau_1^{exc} + \tau_2^{exc})/(e D_{ls}^{crit})$.

FIG. 2: Tunnel resistance (top) and spin signal $\Delta V_{Hanle}$ divided by the current density $J$ (bottom) as a function of the density of localized states $D_{ls}$, for pure two-step tunneling (blue), pure direct tunneling (pink), and for two-step tunneling and direct tunneling in parallel (black - the dotted line is for $\tau_2^{esc}$ and thus $R_2$ reduced by a factor of 1000). The horizontal axis is normalized to the value of $D_{ls}$ for which the currents by direct and two-step tunneling are equal. The top and bottom vertical axes are normalized to, respectively, $R_{tun}$ and the spin signal for pure direct tunneling. The escape times $\tau_1^{esc}$, $\tau_2^{esc}$ as well as $R_{tun}$ were taken to be independent of $D_{ls}$. The inset displays the spin signal versus $R_2$ for $D_{ls} = D_{ls}^{crit}$ and $D_{ls} = 0.01 D_{ls}^{crit}$.
Finally, we discuss an important and often overlooked characteristic of two-step tunneling. The value of \( r_s^{eff} \) (which governs \( \Delta \mu^{ls} \)) can be much larger than \( r_s^{ch} \), but \( r_s^{eff} \) cannot be larger than \( R_2 \). A smaller \( R_2 \) means a stronger coupling between localized states and semiconductor channel, which tends to equalize their spin accumulations and suppress \( \Delta \mu^{ls} \). Hence, any enhancement of the Hanle signal by localized interface states, if present, can be suppressed by reducing \( R_2 \), i.e., by reducing the energy barrier that separates localized states from the semiconductor bulk. For example, when \( \tau_r^{esc} \) and thus \( R_2 \) is reduced by a factor of 1000 at fixed \( D_{ls} \), the maximum Hanle signal is also reduced by about the same factor (Fig. 2, dotted black curve, and inset). Moreover, enhancement becomes limited to a narrower interval around \( D_{ls}^{crit} \). This feature was exploited in the experiments by Dash et al. to exclude interface states as a source of the large spin accumulation observed in silicon at room temperature\(^2\). They used a treatment with Cs to reduce the Schottky barrier, but found spin signals to remain large and much larger than can be supported by the small Schottky barrier (small \( R_2 \)). We suggest that if spin signals are observed that exceed the predictions of spin injection theory, one must look beyond the magnitude of the signal and investigate trends in order to determine whether an enhancement due to localized states is at play. The model presented here describes how two-step tunneling via localized interface states affects the injection and the detection of spin with a ferromagnetic contact, and the resulting trends, providing a firm basis for comparison with experiments.

Appendix A: Currents, potentials and Hanle signal

In this appendix we provide the equations for the current by direct and two-step tunneling for each spin orientation separately. For the sake of completeness, we also provide the full solutions for the potentials and the Hanle signals, without approximations.

For direct tunneling between ferromagnet and semiconductor channel, we denote the tunnel currents of majority (↑) and minority (↓) spin electrons by \( I_\uparrow \) and \( I_\downarrow \), respectively, and the corresponding tunnel conductances by \( G_\uparrow \) and \( G_\downarrow \). With the voltage definitions described in the main text we have:

\[
I_\uparrow = G_\uparrow \left( V - \frac{\Delta \mu^{ch}}{2} \right) \quad (A1)
\]

\[
I_\downarrow = G_\downarrow \left( V + \frac{\Delta \mu^{ch}}{2} \right) \quad (A2)
\]

The charge tunnel current \( I = I_\uparrow + I_\downarrow \) and the spin tunnel current \( I_s = I_\uparrow - I_\downarrow \) due to direct tunneling are then:

\[
I = G V - P_G \frac{\Delta \mu^{ch}}{2} \quad (A3)
\]

\[
I_s = P_G G V - G \frac{\Delta \mu^{ch}}{2} \quad (A4)
\]

with the total conductance \( G = G_\uparrow + G_\downarrow \) and the tunnel spin polarization \( P_G = (G_\uparrow - G_\downarrow)/(G_\uparrow + G_\downarrow) \).

For two-step tunneling via localized interface states, we denote the tunnel currents between ferromagnet and localized states of majority and minority spin electrons by \( I_{1\uparrow} \) and \( I_{1\downarrow} \), respectively, and the corresponding tunnel conductances by \( G_{1\uparrow} \) and \( G_{1\downarrow} \). Tunneling between localized states and semiconductor channel is described by the tunnel currents \( I_{2\uparrow} \) and \( I_{2\downarrow} \), and a tunnel conductance \( G_2/2 \) per spin. The latter is independent of spin because the semiconductor and the localized states are both not ferromagnetic. The tunnel current components for two-step tunneling via localized states are:

\[
I_{1\uparrow} = G_{1\uparrow} \left( V_{ls} - \frac{\Delta \mu^{ls}}{2} \right) \quad (A5)
\]

\[
I_{1\downarrow} = G_{1\downarrow} \left( V_{ls} + \frac{\Delta \mu^{ls}}{2} \right) \quad (A6)
\]

\[
I_{2\uparrow} = \frac{G_2}{2} \left( V - V_{ls} + \frac{\Delta \mu^{ls} - \Delta \mu^{ch}}{2} \right) \quad (A7)
\]

\[
I_{2\downarrow} = \frac{G_2}{2} \left( V - V_{ls} - \frac{\Delta \mu^{ls} - \Delta \mu^{ch}}{2} \right) \quad (A8)
\]
The charge tunnel current $I_1 = I_{1 \uparrow} + I_{1 \downarrow}$ and spin tunnel current $I_{s,1} = I_{s,1 \uparrow} - I_{s,1 \downarrow}$ between ferromagnet and localized states are then:

$$I_1 = G_1 V_{ls} - P_{G1} G_1 \left( \frac{\Delta \mu_{ls}}{2} \right)$$  \hspace{1cm} (A9)

$$I_{s,1} = P_{G1} G_1 V_{ls} - G_1 \left( \frac{\Delta \mu_{ls}}{2} \right)$$  \hspace{1cm} (A10)

with $G_1 = G_1 \uparrow + G_1 \downarrow$ and $P_{G1} = (G_1 \uparrow - G_1 \downarrow) / (G_1 \uparrow + G_1 \downarrow)$. The charge tunnel current $I_2 = I_{2 \uparrow} + I_{2 \downarrow}$ and spin current $I_{s,2} = I_{s,2 \uparrow} - I_{s,2 \downarrow}$ between localized states and semiconductor are:

$$I_2 = G_2 (V - V_{ls})$$  \hspace{1cm} (A11)

$$I_{s,2} = G_2 \left( \frac{\Delta \mu_{ls} - \Delta \mu_{ch}}{2} \right)$$  \hspace{1cm} (A12)

Equations (A3), (A4) and (A9)-(A12) are given in the main text.

The solution for the potential of the localized states is:

$$V_{ls} = \left( \frac{R_1}{R_1 + R_2} \right) \left\{ 1 + \frac{(R_2)^2}{R_1 R_{tun}} \right\} \left\{ \beta_{ch} P_{G1} + P_G \right\} \beta_{ch} \beta_{ls} - 1 \} \} V$$  \hspace{1cm} (A13)

The voltage across the tunnel contact is related to the total current $I^{tot} = I + I_2$ by:

$$V = \frac{R_1 + R_2}{(R_1 + R_2 + R_{tun}) - R_2 \frac{\beta_{ls}(P_G)^2 + \beta_{ch}(P_{G1})^2 + 2 P_G P_{G1}}{\beta_{ch} \beta_{ls} - 1}} R_{tun} I^{tot}$$  \hspace{1cm} (A14)

The full expressions for the Hanle signal in terms of $V$ or $I^{tot}$ are:

$$\Delta V_{Hanle} = \left( \frac{R_2}{R_1 + R_2 + R_{tun}} \right) \left\{ \frac{\beta_{ls}(P_G)^2 + \beta_{ch}(P_{G1})^2 + 2 P_G P_{G1}}{\beta_{ch} \beta_{ls} - 1} \right\} V$$  \hspace{1cm} (A15)

$$\Delta V_{Hanle} = \left\{ \frac{R_2 (R_1 + R_2)}{R_1 + R_2 + R_{tun}} \frac{\beta_{ls} \beta_{ls} - 1}{\beta_{ls} (P_G)^2 + \beta_{ch} (P_{G1})^2 + 2 P_G P_{G1}} - R_2 \right\} R_{tun} I^{tot}$$  \hspace{1cm} (A16)
It was confirmed that eqns. (7) and (8) reduce to the results for pure direct tunneling and pure two-step tunneling in the appropriate limits, respectively, \( R_{\text{tun}} \ll R_1 + R_2 \) and \( R_{\text{tun}} \gg R_1 + R_2 \). Note that setting \( P_G \) (or \( P_{G1} \)) to zero is not sufficient, as this does not remove the charge current and also not the complete spin current.

The term proportional to \( P_G \) in eqn. (11) implies that \( \Delta \mu^{ls} \) is also enhanced by including direct tunneling. This is because it increases \( \Delta \mu^{ch} \), which tends to reduce the spin current \( I_s \) between localized states and the channel since this is proportional to \( \Delta \mu^{ls} - \Delta \mu^{ch} \). The spin accumulation in the localized states then has to increase to restore the balance of the spin currents going in and out. The relative effect is small when \( \Delta \mu^{ls} \gg \Delta \mu^{ch} \).