Theory of spin-polarized current flow through a localized spin triplet state

Stephen R. McMillan and Michael E. Flatté

1Department of Physics and Astronomy, University of Iowa, Iowa City, Iowa 52242, USA
2Department of Applied Physics, Eindhoven University of Technology, Eindhoven, The Netherlands

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We derive a formalism describing quantum-coherent features of spin-polarized charge current through a partially-polarized spin triplet defect in a transverse magnetic field. We predict distinct few-milli-tesla-dc magnetoresistance signatures that identify a single spin-triplet center’s character and reveal the orientation of the spin triplet’s zero-field splitting axis relative to the magnetic contact’s polarization. For example, in 4H-SiC the single (hh), (kk), (hk), and (kh) divacancies are all distinct. Spin-polarized current flow efficiently polarizes the spin, potentially electrically initializing spin-triplet-based qubits.

Spin-based technology relies primarily on the ability to predict and control coherent spin dynamics[1–4]. Efforts to control single solid-state spins have been underway for decades, with defects in semiconducting hosts providing robust and tunable realizations of coherent spin centers[5–7] that can be applied to nanoscale sensing[8], quantum information processing[9] and single photon emission[10]. The capability to deterministically place and identify defects has advanced in parallel[7, 11–17] as a key enabling step for large-scale quantum-coherent systems. For such goals the neutral divacancies in 4H-SiC possess several advantages: long coherence times[18–22], even at room temperature, optical initialization and readout properties similar to NV− centers, emission in the telecom range,[23, 24] and the potential to be manipulated within electrical semiconductor device structures[25, 26]. Prior work used optical techniques to polarize or probe the spin 1 divacancy, followed by manipulation with external fields (including microwave irradiation), however these same operations could in principle be performed entirely electrically on far smaller scales than optical wavelengths. Even without coherent microwave manipulation the spin-coherent nature of spin-polarized transport through defects produces remarkable dc magnetoresistive features, whether through coherent[27, 28] or incoherent[29–31] orbital transport through the defect. However these dc magnetoresistance theories[27, 31] apply to defects with a single orbital state, and a spin transition between spin 1/2 and spin 0 (1/2 ↔ 0). A SiC divacancy undergoes spin 1 ↔ 1/2 transitions during electrical transport, and offers distinct features from spin 1/2 ↔ 0 transitions, such as zero-field splitting of the spin 1 state.

Here we calculate the dc magnetoresistance (MR) of spin-polarized current through a localized spin-triplet state such as a divacancy in 4H-SiC. Our formalism for this dynamical process tracks the spin 1 state (neutral divacancy) with a $4 \times 4$ density matrix $\rho_1$, which must be projected onto the triplet subspace, and the spin 1/2 state (ionized divacancy) with a $2 \times 2$ density matrix $\rho_{1/2}$, along with transitions between the two subspaces mediated by transport processes. The current in a small static (~ millitesla) magnetic field responds sensitively to the relative orientation of the spin-polarized contact.
and the divacancy ZFS axis. The MR emerges from an induced spin polarization of the defect, a pathway to electrically initialize divacancy qubits. The MR signatures also enable sorting divacancy ensembles into their fractional composition of each divacancy configuration.

The steady-state current is obtained from dynamics described by the stochastic Liouville equation,

$$\frac{d\rho(t)}{dt} = -\frac{i}{\hbar} [H, \rho(t)] - D[\rho] + G[\rho],$$

where $D[\rho]$ and $G[\rho]$ represent the dissipators and generators of Fock states that differ by one carrier (spin 1 or spin 1/2) respectively, with $d\rho/dt = 0$. The spin Hamiltonians are, for the spin 1/2 state,

$$H_{1/2} = g\mu_B \mathbf{B} \cdot \mathbf{s},$$

with $\mathbf{s}$ the spin 1/2 operators and $\mathbf{B}$ the applied field, and for the spin 1 state,

$$H_1 = g\mu_B \mathbf{B} \cdot \mathbf{S} + DS_z^s - E(S_x^s - S_y^s),$$

where $\mathbf{S} = \mathbf{s}_a + \mathbf{s}_b$ is the spin 1 operators (only one spin 1/2 is occupied in Eq. (2) so no distinguishing label is needed), and $D$ and $E$ are the longitudinal and transverse zero-field splitting parameters respectively. These two Hamiltonians describe the coherent spin evolution of the density matrices in $\mathbf{B}$ and the ZFS terms.

$\gamma_N$ is the rate for orbitally incoherent hopping from the nonmagnetic bulk to the defect. The generator of the spin 1 state

$$\mathcal{G}_{1/2 \rightarrow 1} = \frac{2}{3} \gamma_N P_1 (\mathbf{I}_{2 \times 2} \otimes \rho_{1/2}) P_1,$$

where $P_1$ projects onto the spin-1 subspace and the factor of $2/3$ normalizes the trace. Generating a spin 1 state also dissipates the spin 1/2 state, according to

$$\mathcal{D}_{1/2 \rightarrow 1} = -\gamma_N \rho_{1/2}.$$  

Dissipation of the neutral spin 1 state and generation of the spin 1/2 ionized state occur via hopping with rate $\gamma_F$ to the spin-selective FM contact with a magnetization operator $\hat{M}$. The anti-commuting form of $\hat{M}$ correctly describes the decay of coherence \cite{footnote}:

$$\mathcal{D}_{1 \rightarrow 2} = \gamma_F \{ \{ \hat{M}^{(a)}, \rho_1 \} \},$$

where $\rho_1 = \rho_a \otimes \rho_b$ and

$$\hat{M}^{(j)} = \frac{1}{4} (\mathbf{I}_{4 \times 4} + \Sigma^{(j)}),$$

with $j = a, b$ corresponding to an individual spin-1/2 subspace, $P$ the in-plane polarization of the FM contact, and $\Sigma^{(j)}$ the $4 \times 4$ Pauli matrix for subspace $j$. For simplicity we assume 100% FM polarization ($P = 1$); lower polarization decreases the MR contrast. The spin 1/2 manifold generation

$$\mathcal{G}_{1 \rightarrow 1/2} = \gamma_F (\text{Tr}_a[\{ \hat{M}^{(a)}, \rho_1 \}] + \text{Tr}_b[\{ \hat{M}^{(b)}, \rho_1 \}]),$$

where $\text{Tr}_j$ is the partial trace over the spin 1/2 subspace $j$. These terms lead to divacancy spin decoherence.

In addition to decoherence from transport, the spin can decohere within a single manifold through interactions with the local environment, characterized by the longitudinal relaxation time, $T_1$, and spin decoherence time $T_2$. For $\rho_{1/2}$ the on-site dissipation is expressed with Lindblad terms:

$$L_1 = \frac{1}{\sqrt{2T_1}} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix},$$

$$L_2 = \frac{1}{\sqrt{2T_2}} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix},$$

$$L_3 = \frac{1}{2T_2} - \frac{1}{4T_2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$

Direct products between two spin 1/2 sub-spaces extends these expressions to the spin 1 manifold with $i = 1, 2, 3$.

$$L_i^{(a)} = P_1 (L_i \otimes \mathbf{I}_{2 \times 2}) P_1,$$

$$L_i^{(b)} = P_1 (\mathbf{I}_{2 \times 2} \otimes L_i) P_1,$$

Putting these expressions together yields:

$$\frac{d\rho_{1/2}(t)}{dt} = -\frac{i}{\hbar} [H_{1/2}, \rho_{1/2}(t)]$$

$$-\mathcal{D}_{1/2 \rightarrow 1}[\rho_{1/2}(t)] + \mathcal{G}_{1 \rightarrow 1/2}[\rho_{1}(t)]$$

$$+ \sum_{j=1}^{3} \left( L_j \rho_{1/2}(t) L_j^+ - \frac{1}{2} \{ L_j^+ L_j, \rho_{1/2}(t) \} \right)$$

$$\frac{d\rho_1(t)}{dt} = -\frac{i}{\hbar} [H_1, \rho_1(t)] - \mathcal{D}_{1 \rightarrow 1/2}[\rho_{1}(t)]$$

$$+ \mathcal{D}_{1/2 \rightarrow 1}[\rho_{1/2}(t)]$$

$$+ \sum_{j=1}^{3} \left( L_{j}^{(a)} \rho_1(t) L_{j}^{(a) \dagger} - \frac{1}{2} \{ L_{j}^{(a) \dagger} L_{j}^{(a)}, \rho_1(t) \} \right)$$

$$+ \sum_{j=1}^{3} \left( L_{j}^{(b)} \rho_1(t) L_{j}^{(b) \dagger} - \frac{1}{2} \{ L_{j}^{(b) \dagger} L_{j}^{(b)}, \rho_1(t) \} \right),$$

where $\{ ... \}$ represents anti-commutation.

The operator corresponding to current onto the defect is proportional to $\mathcal{G}_{1 \rightarrow 1/2}$, and off of the defect is proportional to $\mathcal{D}_{1 \rightarrow 1/2}$. They are

$$\hat{I}_N = e \mathcal{G}_{1 \rightarrow 1/2} = \frac{2e\gamma_N}{3} P_1 (\mathbf{I}_{2 \times 2} \otimes \rho_{1/2}) P_1,$$

$$\hat{I}_F = e \mathcal{D}_{1 \rightarrow 1/2} = e\gamma_F \{ \{ \hat{M}^{(a)}, \rho_1 \} \}. $$

The charge current is calculated from the trace of these operators, $I = \text{Tr}\hat{I}_N = \text{Tr}\hat{I}_F$. 

We now apply this general formalism to \((hh)\) and \((kk)\) divacancies in 4H-SiC with the \(c\) axis normal to the surface [Fig. 1(a)]. These divacancies are \(C_{3v}\) symmetric with defect axis along the \(c\)-axis of the crystal, and as a result the crystal field does not induce a transverse zero-field splitting. The applied bias also will not contribute to a transverse zero-field term, and thus \(E = 0\) in Eq. (3). As depicted in Fig. 1(a), we consider \(\mathbf{B} \parallel c\) axis and the contact magnetization \(\perp c\) axis. In this high-symmetry configuration the current through an individual divacancy can be calculated analytically, and in the limit \(\gamma_N \to \infty\),

\[
I_{\gamma_N \to \infty}(\mathbf{B}) = e\gamma_F \frac{1728(\mathbf{B}^3 - \mathbf{B}D^2)^2 + 48(50\mathbf{B}^4 - 3\mathbf{B}^2D^2 + 9\mathbf{D}^2)\gamma_F^2 + 75(4\mathbf{B}^2 + 3\mathbf{D}^2)\gamma_F^4}{1728(B^3 - BD^2)^2 + 16(166B^4 + 39B^2D^2 + 27D^4)\gamma_F^2 + (700B^2 + 417D^2)\gamma_F^4 + 100\gamma_F^6},
\]

where \(\mathbf{B} = g\mu_B B/\hbar\) and \(\mathbf{D} = D/\hbar\).

![Figure 2](image)

**FIG. 2.** (a) MR for \((hh)\) divacancies with azimuthal angle of 0 (orange), \(2\pi/3\) (blue), and \(4\pi/3\) (green) relative to the FM magnetization. (b) MR for \((kk)\) divacancy with azimuthal angle of \(\pi/3\) (blue), \(\pi\) (orange), and \(5\pi/3\) (green). For both \(\gamma_F/\gamma_N = 0.02\). Inset: sketch of \((hh)\) and \((kk)\) divacancy orientation and the FM magnetization viewed along the \(c\)-axis.

Current from the bulk 4H-SiC substrate through the defect will respond to an applied magnetic field due to the non-equilibrium spin-spin correlation between the magnetization of the planar contact and the spin state of the defect. The finite-field features in Fig. 1(d) reflect the induced polarization of the defect spin as a result of these non-equilibrium correlations.
FIG. 3. Ensemble magnetoresistance for a uniform distribution of \((kh)\) (a), \((hk)\) (b), and each of the four (c) divacancies for two different hopping ratios: \(\gamma_F/\gamma_N = 0.02\) (blue) and \(\gamma_F/\gamma_N = 0.002\) (orange).

FIG. 4. (a) Spin polarization of \((hh)\) divacancy. (Inset) contour plot of \(B > 0\) total spin polarization, with light color indicating a maximum. Points of interest are labeled in the defect orientation basis. \(\hat{z} \parallel \) defect axis and \(\perp\) to the interface, with FM polarization \(\parallel \hat{x}\). The maximum polarization is 0.3 along the \((\theta, \phi) = (1.11, 3.26)\) direction. (b) Positive field current dip of \((hh)\) divacancy with negligible (orange, \(T_2 \gamma_N = 10^8\)), considerable (blue, \(T_2 \gamma_N = 100\)), and substantial (green, \(T_2 \gamma_N = 10\)) on-site decoherence, assuming three different \(T_1\)’s. Decoherence broadens the signal whereas finite \(T_1\) shifts to smaller \(B\). \(\gamma_F/\gamma_N = 0.02\).

the MR features for defects in identical crystal environments. Conversely, differences in the zero-field splitting parameters provide a means of identifying the local crystal environment through measurement of the finite-field current dips. The scale of the defect spin polarization and the degree of asymmetry depend on the relative orientation of the defect axis, providing a distinct signature for each unique orientation of a divacancy.

A mixed ensemble of non-interacting divacancies exhibits MR with increased complexity due to each distinct contributing signature. Interacting defects are not considered here, but the example of two spin-1/2 centers interacting through exchange has been shown to modify the magnetoresistance. The results described here describe ensembles with typical defect spacings \(\sim 10\) nm. Dipolar interactions at this distance are of the order of 500 kHz and exchange interactions even smaller — unresolvable for the currents considered here. The MR thus directly provides the concentration of contributing defects of a given orientation. The total current \(I(B)_{\text{comp}} = \sum_i w_i I_i(B)\), where \(w_i\) indicates the fractional population and \(i\) labels divacancy configuration.

Figure 3 shows the composite signal for \((kh)\): \(w(hk)_0 \approx w(hk)_2/3 = w(hk)_4/3 = 1/3\), \((hk)\): \(w(hk), \pi/3 = w(hk), 5\pi/3 = 1/3\), and a uniform mixture of the four types of divacancies \((wi = 1/8)\) respectively. The two traces represent hopping ratios that differ by an order of magnitude. For \(\gamma_N \approx \text{ns}^{-1}\), the applied magnetic field required to resolve the zero-field energies is \(\sim\) mT. The smallest energy scale of the system is set by \(\gamma_F\); in Fig. 3 the higher resolution is obtained when \(\gamma_F \approx 2\) MHz, corresponding to single-defect currents of 0.3 pA and \(T_2 > 10\) ms. The total current scales with the defect number, and such \(T_2\)’s are typical for divacancies in 4H-SiC.

Divacancy spin decoherence further influences the features discussed above. Figure 4(a) shows the spin for an \((hh)\) divacancy. The FM polarization \(\parallel \hat{x}\), \(\mathbf{B} \parallel \hat{z}\) (the defect axis). At the current dip the spin polarization is
primarily in the $xz$ plane, indicating an axis of maximum polarization oblique to the defect axis. The inset of Fig. 4(a) is a contour plot of the total spin polarization at the positive $B$ field current dip in the defect basis. The $+\hat{x}$ and $+\hat{z}$ directions are red and the $+\hat{y}$ is black. Lighter colors indicate positive values and darker colors indicate negative values. The axis of maximum polarization has an orientation of $(\theta, \phi) = (1.11, 3.26)$ radians in the $xz$ basis with a magnitude of $|S_{\text{max}}| = 0.3$. 

Fig. 4(b) shows the steady-state current dip at $+B$ for a single (hh) defect from Eq. (2)'s solution. We consider three different regimes of operation: weak ($T_2^* \gamma = 10^5$, orange), moderate ($T_2^* \gamma = 10^2$, blue), and strong ($T_2^* \gamma = 10$, green) dephasing, with a fixed hopping ratio $\gamma / \gamma = 0.02$. In each regime we consider three relationships between the longitudinal relaxation $T_1$ and the decoherence time $T_2$ indicated by the line type. In the weak regime, all three relations are equivalent and the feature reduces to the trace for negligible on-site mixing. In the moderate and strong regimes show a general broadening of the feature, with $T_1$ shifting the feature to lower field, as is most apparent in the strong regime.

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