Temperature dependent dynamical nuclear polarization bistabilities in double quantum dots in the spin-blockade regime

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(Dated: May 5, 2014)

The interplay of dynamical nuclear polarization (DNP) and leakage current through a double quantum dot in the spin-blockade regime is analyzed. A finite DNP is built up due to a competition between hyperfine (HF) spin-flip transitions and another inelastic escape mechanism from the triplets, which block transport. We focus on the temperature dependence of the DNP for zero energy-detuning (i.e. equal electrostatic energy of one electron in each dot and a singlet in the right dot). Our main result is the existence of a transition temperature, below which the DNP is bistable, so a hysteretic leakage current versus external magnetic field 𝐵 appears. This is studied in two cases: (i) Close to the crossing of the three triplet energy levels near 𝐵 = 0, where spin-blockade is lifted due to the inhomogeneity of the effective magnetic field from the nuclei. (ii) At higher 𝐵-fields, where the two spin-polarized triplets simultaneously cross two different singlet energy levels. We develop simplified models leading to different transition temperatures 𝑇c, ⋆ and 𝑇c, ⋆ for the crossing of the triplet levels and the singlet-triplet level crossings, respectively. We find 𝑇c, ⋆ analytically to be given solely by the HF couplings, whereas 𝑇c, ⋆ depends on various parameters and 𝑇c, ⋆ > 𝑇c, ⋆ .

The key idea behind the existence of the transition temperatures at zero energy-detuning is the suppression of energy absorption compared to emission in the inelastic HF transitions. Finally, by comparing the rate equation results with Monte Carlo simulations, we discuss the importance of having both HF interaction and another escape mechanism from the triplets to induce a finite DNP.

I. INTRODUCTION

The high degree of experimental control in modern quantum dot systems allows detailed manipulation of electrons and their spin in confined states. A particularly intriguing example was investigated by Ono et al.5,6 in a series of experiments. These revealed that, not only the charge, but also the Pauli exclusion principle for spin states can block the electronic transport through a double quantum dot (DQD) coupled in series. To observe this phenomenon – dubbed spin-blockade (SB) – the energy levels of the two dots are tuned asymmetrically, so an electron with a definite spin is trapped in –say– the right dot. Now, only electrons with the opposite spin (to the trapped one) can pass through the DQD, since two electrons with equal spins in the right dot is tuned to be energetically forbidden. Therefore, once an electron with a definite spin is trapped in –say– the right dot, then transport through the DQD is blocked. SB requires non-linear bias and due to the asymmetric energy level tuning of the dots, current is only blocked in one direction leading to the observed current rectification.3

An electron can escape from the states blocking transport by a spin relaxation process, which leads to a small leakage current in the SB regime. Analyzing the leakage current is therefore an excellent tool to obtain information about the spin relaxation processes from a transport experiment. There are several ways to escape from the blocking states: via co-tunneling processes, spin-orbit mediated spin relaxation, and/or by hyperfine interaction (HFI) between the electronic spins and the nuclear spins of the host material. The relative importance of these mechanisms depends on the material and the external parameters. For instance, a specific co-tunneling process can become important by tuning the gate-voltages such that the virtual energy exchange in the co-tunneling process becomes low.

Ever since the experiments by Ono et al.5,6 in vertical GaAs DQDs, several geometries and materials have been used to further study the leakage current in the SB regime, due to different relaxation mechanisms. For instance, Churchill et al.23 analyzed experimentally the leakage current in carbon nanotube DQDs varying the amount of 13C — the only stable carbon isotope with a non-zero spin. This amounts to varying the spin relaxation due to HFI from very important (high 13C concentration) to not important (almost no 13C present). This shows how different the leakage current can be with and without nuclear spins. Nowadays, spin-orbit coupling is also thought to play a role in carbon nanotubes.24

SB in Silicon DQDs has also been studied.25 In a recent work, Lai et al.25 eliminated the HFI in Silicon DQDs by isotopic purification — along the same lines as Churchill et al. In this case, co-tunneling processes caused the leakage current in the SB regime (in good agreement with recent theories), since the spin-orbit coupling is expected to be weak in Silicon. In contrast, spin-orbit interaction is generally believed to be strong in InAs. This enabled Pfund et al.25 to investigate its importance on the leakage current in InAs nanowire DQDs. Finally, recent studies show bipolar SB triple dot and...
valley-spin blockade in carbon nanotube DQDs.  

A HF-induced spin relaxation process from a blocking state will flip the electronic and nuclear spin in opposite directions, Fig. 1(a). The electronic spin relaxation in the SB regime can therefore change the average occupations of the nuclear spin states, since the nuclear spin relaxation time is very long compared to the electronic tunneling timescales. The repeated electronic spin-flip due to the leakage current can therefore produce a dynamical nuclear polarization (DNP). The DNP acts on the electronic states of the DQD as an effective magnetic field, the so-called Overhauser field. The Overhauser field is generally inhomogeneous and therefore often different in the two dots. DNP is also studied in optical and quantum Hall systems.

It is important to emphasize that even though the HF-induced spin-relaxation flip do flip a nuclear spin, it will not always change the average polarization in the steady state of the SB regime. For instance, if HFI is the only spin relaxation mechanism causing the leakage current, then the nuclear spins remain unpolarized. Essentially, this is because tunneling into one of the two blocking states consisting of two spin-up electrons, $|\uparrow\uparrow\rangle$, or two spin-down electrons, $|\downarrow\downarrow\rangle$, is equal. Escape from these two states will polarize the nuclei in opposite directions and therefore on average the polarization does not change. This is so, even though the escape rates from the blocking states might be very different. In this case of HFI being the only cause of leakage current, the nuclear spins can be modeled as an effective magnetic field with zero mean value and non-zero statistical deviation, as has also been used to fit experimental data. Nevertheless, if more than one spin relaxation mechanism contribute to the leakage current in the SB regime, then the nuclei can indeed obtain a non-zero DNP.

The finite DNP leads to experimentally measurable signatures in the leakage current. Perhaps the most fascinating of these signatures is that of a hysteretic current versus external parameters like the magnetic field or a gate-voltage as has been observed. For instance, Pfund et al. found hysteresis due to a competition between HF and spin-orbit induced escape from the blocking states. The hysteresis signals a bistability in the DNP: For a certain range of parameters, there exists two stable values of the DNP leading to two values of the current. For other parameters the nuclear spins might be polarized, but the DNP is single-valued and hence also the current.

Very high polarizations of about half of the nuclei have been found experimentally in the SB regime and even higher DNP were not excluded. Spin diffusion from the DQD to the environment and dipole-dipole interactions are very weak, but nevertheless expected to reduce the polarizations somewhat.

Other experimental findings like long-lived current oscillations in time and transient phenomena in the leakage current have also been attributed to the nuclear spin environment. Furthermore, it has been shown that DNP can build up in DQDs by cycles in gate-voltage space – without transport through the DQD.

The HFI is most effective to lift SB close to the crossing of the electronic energy levels between e.g. a triplet and the singlet state such that energy is conserved in the spin-flip process. In order to get close to a level crossings, the local gate voltages on the dots, the inter-dot tunneling or the external magnetic field can be varied experimentally. The local gate voltages change the energy levels of the individual dots and thereby the so-called energy detuning (i.e. the electrostatic energy difference between one electron in each dot and a singlet in the right dot). The barrier between the dots controls the wavefunction overlap and therefore the quantum mechanical exchange energy between the singlet and triplet states. Moreover, the external magnetic field splits up the triplet levels.

A. Main ideas of this work and comparison to previous works

In this work, we analyze the leakage current and the DNP in the SB regime. Finite DNP occurs due to a competition between (i) the HF-induced escape from the blocking states and (ii) another weaker inelastic escape.

**FIG. 1.** (Color online) (a) Illustration of the two escape paths from the blocking triplets $T$, which compete to create a non-zero DNP in the SB regime. The cycle of transporting an electron through a blocking triplet state $T$ begins and ends with a single electron trapped in the right dot $0(0,1)$. During a single transport cycle, the DNP $P$ is changed by a small amount $\pm dP$ (depending on the specific transition), if the escape from $T$ is HF mediated (upper branch). The DNP acts back on the energy levels $E(P)$ via the Overhauser fields and, in turn, on both the HF spin-flip rates $W$ and the tunneling rates $\Gamma$. In contrast, the weak inelastic escape mechanism (lower branch) leaves the DNP unchanged. (b) A real-space example of a HF-induced escape process from a triplet to the right contact through a singlet $S$. This changes the DNP.
mechanism such as co-tunneling or spin-orbit interaction – as in the works by Rudner et al.\textsuperscript{[25–26]} The induced DNP acts back on the electronic energy levels of the DQD, which in turn change the transition rates until the steady state is reached. Hence, we are dealing with a non-linear system with feedback present as illustrated in Fig. 1.

We consider the gate-voltage configuration with zero energy detuning, i.e. the electrostatic energy of one electron in each dot and a singlet in the right dot is the same. Thus, the external magnetic field is varied to get close to level crossings. The Overhauser magnetic field from the DNP is different in each dot, which mixes the triplet and singlet states with zero total angular momentum projection.\textsuperscript{[27–30]} Thus, we study two kinds of level crossings in detail: The crossing of the triplet (like) levels and the crossing between singlet and triplet levels. What we name the crossing of triplet levels is in fact the crossing of the pure spin-polarized triplets and the triplet that has a small mixing with the singlet subspace. Hence, escape from the mixed state is possible.

The main focus of this work is the presence and description of a transition temperature $T_c$ for the DNP. For temperatures $T$ below $T_c$, the leakage current shows hysteresis versus the external magnetic field $B$, while for $T > T_c$, the hysteresis disappears even though the system can still have a non-zero DNP. The transition temperature is related to a bistability of the DNP for $T < T_c$, which is the reason for the current hysteresis. Interestingly, we find that the transition temperatures for the crossing of the triplet levels near $B = 0$, $T_{c,TT}$, and the singlet-triplet crossing at finite $B$-field, $T_{c,ST}$, respectively, are in general different and $T_{c,TT} < T_{c,ST}$ for typical parameters. Thus, for $T_{c,TT} < T < T_{c,ST}$ current hysteresis is expected near the singlet-triplet level crossings at finite $B$-field, but not near the crossing of the triplet levels.

The DNP in the SB regime is current induced and, hence, a result of a non-equilibrium situation. Remarkably, spontaneous order of the nuclear spins in equilibrium generally happens at orders of magnitude lower temperatures than $T_{c,TT}$ and $T_{c,ST}$ due to the weakness of dipole-dipole interaction among the nuclear spins.\textsuperscript{[29,13–102]}

We find the transition temperature $T_{c,TT}$ analytically to be given only by the strength of the HF couplings in the DQD. This is derived from a simplified model valid in the limit of the singlets being far away from energy from the triplets, i.e. at large exchange energy. To describe the singlet-triplet crossing, we also derive a simplified model leading to an implicit equation for the DNP. In contrast to $T_{c,TT}$, we find the transition temperature $T_{c,ST}$ for the singlet-triplet crossing to depend on various parameters.

The possibility of not conserving energy in the HF transitions is present in this work. Rudner et al.\textsuperscript{[25–26]} include this effect as level broadening, whereas we allow for energy emission and absorption e.g. by phonons in the HF rates. Hence, energy absorption and emission in a HF process is equally likely in Refs.\textsuperscript{[25–26]}. In contrast, here the probability for energy absorption is exponentially suppressed compared to energy emission.\textsuperscript{[116–118]}

We show that this is indeed an essential difference between this study and the previous ones,\textsuperscript{[25–26]} since the presence of both transition temperatures exactly stem from this asymmetry between energy emission and absorption.

In previous works by some of us,\textsuperscript{[29–30]} non-zero DNP in the SB regime arise due to the competition between HF-induced spin-flips and escape from the blocking states by tunneling through excited states in the right dot. In contrast, such excited states are assumed to be far away in energy in this work and, hence, do not play a role. Moreover, we work with coherently coupled dots such that the inter-dot tunneling is not treated as a perturbation as in Refs.\textsuperscript{[37–39]}. This approach, for instance, includes the expected triplet with zero angular momentum projection. Moreover, previous\textsuperscript{[29–30]} phonon absorption processes were neglected, so the physics treated here regarding the transition temperature was missed.

Some of us have numerically studied a similar approach recently.\textsuperscript{[22]} However, in this case, the rate equation for the DNP turned out to be inappropriate, because HF1 was taken to be the only escape mechanism from the blocking states. In contrast, here we find DNP to appear due to a competition between HF-induced escape and another inelastic escape mechanism. Here we put our results from the rate equation approach on a firm basis by comparing to Monte Carlo simulations. Furthermore, we point out in detail how the rate equation approach becomes sensitive to some initial occupations in the case without an inelastic escape path, and therefore become unable to describe the physical setup. Moreover, we emphasize that this work contains many new insights and results compared to Ref.\textsuperscript{[22]}. For instance, the simple analytical models describing the various level crossings, which lead to the transition temperatures described above.

The paper is organized as follows: Sec. II describes the model of the DQD energy levels and their interplay with the DNP. Then we address the crossing of the triplets (Sec. III) and the singlet-triplet crossings (Sec. IV). Finally, the Monte Carlo simulations are discussed (Sec. V).

II. THE MODEL

The model used below aims at describing the basic physics of a DQD coupled to a nuclear environment in the SB regime – instead of focusing on a specific material.

A. The states of the DQD, the Hamiltonian and the Overhauser field

The three triplet states blocking the transport in the SB regime are

$$|T_+⟩ = d_L^d_R^† |0⟩, \quad |T_-⟩ = d_L^†d_R |0⟩, \quad |T_0⟩ = \frac{1}{\sqrt{2}} (d_L^d_R^† |0⟩ + d_L^†d_R^† |0⟩),$$

(1a)

In contrast, here the probability for energy absorption is exponentially suppressed compared to energy emission.\textsuperscript{[116–118]}
where the indices 0 and ± represent the total angular momentum projection, m = 0, ±1. The singlet states with one electron in each dot, |S(1, 1)⟩, and two electrons in the right dot, |S(0, 2)⟩, respectively, are

\[ |S(1, 1)⟩ = \frac{1}{\sqrt{2}} (d_{1L}^+d_{1R}^+ - d_{1L}^-d_{1R}^-) |0⟩, \]

\[ |S(0, 2)⟩ = d_{1L}^+d_{1R}^+ |0⟩. \]

A single electron with spin σ trapped in the right dot is described by the one-electron state |σ(0, 1)⟩ = d_{1R}^+ |0⟩ for σ = ↑, ↓. Here we only include a single spin-degenerate state created (annihilated) by \(d_{1L}^+, (d_{1L}^-)\) in the right (α = L) dot. The empty state is |0⟩.

The entire system is described by the Hamiltonian

\[ H = H_{\text{DQD}} + H_{\text{leads}} + H_T + H_{\text{HF}}, \]

where \(H_{\text{DQD}}, H_{\text{leads}}\) and \(H_T\) describe the two dots in series, the electronic leads and the tunneling coupling between them, respectively. The HFI, \(H_{\text{HF}}\), is between the electrons in the DQD and the nuclear spins. The DQD is described by an Anderson-type Hamiltonian

\[ H_{\text{DQD}} = \sum_{\alpha=L,R} (ε_\alpha n_\alpha + U_\alpha n_\alpha n_{\alpha \uparrow} + g_\mu_B BS_{z,\alpha} + U_{RL\sigma} n_\alpha n_{\alpha \sigma} + t_{\sigma} (d_{\alpha \sigma}^+d_{\alpha \sigma}^+) + U_{\alpha} n_\alpha n_{\alpha \uparrow} + g_\mu_B BS_{z,\alpha} + U_{RL\sigma} n_\alpha n_{\alpha \sigma} + t_{\sigma} (d_{\alpha \sigma}^+d_{\alpha \sigma}^+) \). \]

where the nuclear spin polarization \(I_\alpha\) can be read off the Overhauser magnetic field, \(B_{\text{HF}}\), which is crucial for lifting the SB singlet-triplet splitting. The polarization of the nuclei acts back on the electronic states as an effective Overhauser magnetic field \(\vec{B}_{\text{HF}}\).

To include this, we divide the HF Hamiltonian into a mean-field part \(H_{\text{HF}}^{\text{MF}}\), and the Overhauser field, and a spin-flip part \(H_{\text{HF}}^{\text{MF}}\), which leads to the HF-induced spin-flips necessary for dynamically polarizing the nuclei. The external magnetic field provides a direction along which the nuclei can polarize\(^{[10]}\) such that the rotational symmetry is broken as in the experimental situation\(^{[13]}\) for spin-flip. Hence, the mean-field from the nuclei is taken to be along the \(z\)-direction as the external magnetic field and given in terms of the average number of spin up and down \(N_\sigma (\sigma = \uparrow, \downarrow)\) in the nuclear environment\(^{[11]}\). Thus,

\[ H_{\text{HF}}^{\text{MF}} = \sum_{\alpha=L,R} \frac{1}{2} A_\alpha P_S z,\alpha, \]

where the nuclear spin polarization \(P \equiv (N_\uparrow - N_\downarrow)/N\) was introduced and \(N = N_\uparrow + N_\downarrow\) is the total number of nuclear spins. The number of nuclear spin-up (down) \(N_\uparrow (N_\downarrow)\) and thus \(P\) change dynamically according to the external conditions of the current. The spin-flip part of the HFI is

\[ H_{\text{HF}}^{\text{MF}} = \sum_{\alpha=L,R} \frac{A_\alpha}{2N} (S_{\downarrow,\alpha} I_{+\alpha} + S_{\uparrow,\alpha} I_{-\alpha}), \]

where \(S_{\downarrow,\alpha} = S_{\uparrow,\alpha} = iS_{y,\alpha}\) and \(I_{\pm,\alpha} = I_{\pm,\alpha} \pm iI_{y,\alpha}\) are the raising and lowering operators of the electronic and nuclear spins, respectively. The electronic spin-flips induced by \(H_{\text{HF}}^{\text{MF}}\) are included perturbatively below (see Sec. II B).

From \(H_{\text{HF}}^{\text{MF}}\) we can read off the Overhauser magnetic field in each dot \(B_{\text{HF}}^{i\alpha} = \frac{A_\alpha P}{2g_\mu_B}\). Importantly, the Overhauser fields in the two dots are different, \(B_{\text{HF}}^{i\alpha} \neq B_{\text{HF}}^{i\beta}\), which is crucial for lifting the SB singlet-triplet splitting (as will be clear below). Here we introduce this difference by having \(A_L \neq A_R\), but keep the DNP \(P\) as a common quantity for both dots\(^{[11]}\). In principle, the DNP can be spatially inhomogeneous, which is challenging to model in detail. A step on that way, is having different – but homogeneous – polarizations in the two dots and \(A_L = A_R\). However, for coherently coupled dots, the overlap between the envelope functions is sizeable and thereby also the amount of nuclei under both envelope functions. This makes it less clear how to separate the nuclei into forming two independent homogeneous polarizations\(^{[11]}\). For simplicity, we therefore use a single DNP for both dots.

We find the basis states of the DQD including the Overhauser fields and the inter-dot tunneling by diagonalizing \(H_{\text{DQD}} + H_{\text{HF}}^{\text{MF}}\) within the space of triplets \(|T_m⟩\)\(^{[11]}\), singlets \(|S⟩\) and one-electron states \(|σ(0, 1)⟩\), since all other states are not energetically relevant in the SB regime\(^{[11]}\). We specialize to the zero detuning limit such that the electrostatic energy of one electron in each dot,
\[
\varepsilon_L + \varepsilon_R + U_{RL}, \text{ and of a singlet in the right dot, } 2\varepsilon_R + U_R, \text{ are the same: } \\
\varepsilon_L + \varepsilon_R + U_{RL} = 2\varepsilon_R + U_R = 0 \text{ (chosen as the zero of energy). For zero detuning, the diagonalization gives the following particularly simple basis states } \\
|T_+\rangle = d_{L}^{\dagger}d_{R}^{\dagger}|0\rangle, \\
|T_{p}\rangle = \frac{1}{\sqrt{N}}\left(|T_{0}\rangle - p|S(0, 2)\rangle\right), \\
|S_{\pm}\rangle = \frac{1}{\sqrt{2}}\left(|S(1, 1)\rangle \pm \frac{1}{\sqrt{2}}\left(|S(0, 2)\rangle + p|T_{0}\rangle\right)\right),
\]
and the one-electron states remain the same, \(|\sigma(0, 1)\rangle = d_{R}^{\dagger}|0\rangle\) and we set \(\hbar = 1\). Here we introduced
\[
p \equiv \frac{A_{L} - P}{2\sqrt{2}t}, \quad N \equiv \sqrt{1 + p^2}, \quad A_\pm \equiv \frac{A_{L} \pm A_{R}}{2}
\]
and the energies are found to be (see Fig. 2)
\[
E_{T_{0}} = 0, \quad E_{T_{p}} = \pm \left(g\mu_B B + \frac{1}{2} A_+ P\right), \quad E_{S_{\pm}} = \pm \sqrt{2}tN.
\]
Here the inter-dot tunneling mixes \(|S(0, 2)\rangle\) and \(|S(1, 1)\rangle\). Moreover, these two singlets mix with \(T_0\) due to the difference of the Overhauser fields between the two dots \(g\mu_B B_{\text{nuc}} - B_{\text{R}}\) = \(-P/2\). The triplet-singlet mixing is controlled by the dimensionless parameter \(p\) in Eq. 7. The sum of the Overhauser fields, \(g\mu_B (B_{\text{nuc}} - B_{\text{R}}) = A_+ P\), splits the spin-polarized triplets \(T_{\pm}\) as a magnetic field does. Hence, \(E_{T_{\pm}}\) depend stronger on \(P\) than the exchange energy splitting \(|E_{S_{\pm}} - E_{T_{\pm}}| = \sqrt{2}tN\) for \(p \ll 1\). The singlet-triplet mixing \(p\) is indeed small, since we are interested in the limit \(A_- \ll t\). Therefore, we keep calling the state \(T_{p}\) for a triplet and the states \(S_{\pm}\) for singlets (as indicated by the notation), even though strictly speaking they are not.

In the SB regime, the triplets \(T_{p}\) and \(T_{0}\) block transport. Due to the singlet-triplet mixing, the state \(T_{p}\) is not a blocking state anymore, whereas \(T_{\pm}\) still block transport. Therefore, if \(P = 0\) or \(A_{L} = A_{R}\), then \(p = 0\) leading to transport blocking by all three triplets (\(T_{p}\) to \(T_{0}\)).

In contrast to finite detuning \(|\varepsilon_{L} - \varepsilon_{R}|\), the crossing of the singlet and triplet energies always happens in pairs for zero detuning, e.g. \(E_{T_{+}}\) and \(E_{S_{+}}\) cross if and only if \(E_{T_{-}}\) and \(E_{S_{-}}\) cross. Moreover, here the energy levels relative differences have certain symmetries around \(E_{T_{p}}\):
\[
E_{T_{p}} = E_{T_{0}} - E_{T_{p}} = E_{T_{-}} - E_{T_{0}} = E_{S_{+}} - E_{S_{-}}.
\]

B. The dynamical coupling of the nuclear spins to the DQD energy levels and the leakage current

The dynamics is determined within the rate equation approach written compactly as
\[
\begin{align*}
\dot{n}_{\nu} &= \sum_{\nu'} \left(W_{\nu,\nu'} + \Gamma_{\nu,\nu'}^{\text{inel}} \right) n_{\nu'} - n_{\nu} \sum_{\nu'} \left(W_{\nu',\nu} + \Gamma_{\nu',\nu}^{\text{inel}} \right),
\end{align*}
\]
and the time derivative of the average occupation \(n_{\nu}\) for \(\nu = T_{+}, T_{p}, S_{\pm}\) is indeed small, since we are interested in the limit \(t \rightarrow 0\). Moreover, the normalization condition is \(\sum_{\nu} n_{\nu} = 1\). We include three kinds of rates: (i) The HF spin-flip rates \(W_{\nu,\nu'}\), the initial condition is \(n_{\nu} = 0\). (ii) The tunneling rates \(\Gamma_{\nu,\nu'}\), which connect the leads to the DQD. (iii) Finally, we include another inelastic escape rate \(\Gamma_{\nu,\nu}^{\text{inel}}\), which allows for escape from the triplets \(T_{\pm}\) and \(T_{p}\). Sec. II C gives the detailed rates. Fig. 3 provides an illustration of the non-zero rates in the rate equations, which are given explicitly in Appendix A for completeness.

In order to obtain non-zero DNP, a preferred direction of angular momentum transfer from the electrons to the nuclei needs to exist. If HF is the only source of SB lifting, then no such preferred direction exists. The reason is that HF-induced escape from the spin-polarized triplets \(T_{\pm}\) changes the DNP in opposite directions and since the probabilities of loading \(T_{p}\) and \(T_{+}\) are equal \((\Gamma_{T_{+}T_{p}} = \Gamma_{T_{p}T_{+}})\), no net DNP build up. This is so, even though the HF rates from \(T_{p}\) might be very different, but since only one way to escape from \(T_{p}\) exists, it does not matter if escaping from \(T_{p}\) or \(T_{+}\) is the fastest.

Here we allow two ways to escape from the triplets, both lifting SB. Either by the HF spin-flip transitions \(W_{\nu,\nu'}\) or by the inelastic escape rates \(\Gamma_{\nu,\nu'}^{\text{inel}}\). The additional inelastic rates can e.g. be provided by co-tunneling or spin-orbit mediated spin relaxation processes. Importantly, the inelastic processes contained in \(\Gamma_{\nu,\nu}^{\text{inel}}\) give an additional escape path from the triplets without a nuclear spin-flip. Therefore, the two escape paths from the blocking states now compete, such that it becomes important which of the HF-induced escape paths from \(T_{p}\) or \(T_{+}\) is the fastest. This competition is therefore crucial to obtain non-zero DNP. We do not specify the inelastic escape rate further as Rudner et al. 2019.
The transition rates connecting the energy levels of the DQD

![Diagram of transition rates](image)

FIG. 3. (Color online) Illustration of the transitions connecting the DQD states. Each HF transition $W_{f,i}$ flips the electronic and nuclear spins in opposite directions and change the DNP by $dP = \pm 2/N$ or $dP = -2/N$ depending on the transition – as specified in the label of the figure. We allow for emission or absorption of energy e.g. as a phonon (ph.) in the HF processes. Thus, transitions between misaligned energy levels are possible, but they become less probable the larger the difference between the energy levels. Moreover, as discussed in the main text, absorption of energy is suppressed by a factor of $e^{-2\Delta E/k_BT}$ compared to emission, where $\Delta E > 0$ is the difference between the levels. Additional inelastic escape rates $\Gamma_{\sigma,T}^{\text{ine}}$ from the triplets without a nuclear spin-flip are also included (red arrows). The competition between these rates and the HF rates creates the possibility of finite DNP.

Each HF-induced electronic spin-flip will change the DNP by $dP = \pm 2/N$ depending on the transition, see Fig. 3. Therefore, in the case of competing escape rates, we describe the DNP, $P$, by the rate equation

$$\dot{P} = \frac{2}{N} \left[ \left( W_{T_-,S_+} - W_{T_+,S_-} \right) n_{S_+} + \left( W_{T_-,S_-} - W_{T_+,S_+} \right) n_{S_-} + \left( W_{S_+,T_+} + W_{S_-,T_-} - W_{T_+,T_-} \right) n_{T_+} - \left( W_{S_+,T_-} + W_{S_-,T_+} + W_{T_+,T_-} \right) n_{T_-} \right]$$

as used in several other studies. The idea behind Eq. (10) is to describe a competition between various rates that polarize the nuclear spins in opposite directions, which is different from the aforementioned competition between various escape paths. However, in order to have a competition between various rates that polarize the nuclei in the first place, it is strictly necessary to have competing escape paths, i.e. both $W_{f,i}$ and $\Gamma_{\sigma,i}^{\text{ine}}$ non-zero. In Sec. IV we show that the rate equation approach (9,10) fails for $\Gamma_{\sigma,i}^{\text{ine}} = 0$. Moreover, we validate the rate equation approach for $\Gamma_{\sigma,i}^{\text{ine}} \neq 0$, by showing that it leads to the same results as a Monte Carlo simulation.

Furthermore, we neglect processes leading to depolarization of the nuclear bath in equilibrium since these are much slower than the HF spin-flip processes during transport through the DQD in the SB regime.

In this work, we use the high bias limit, where electrons only enter the DQD from the left lead and leave it to the right lead. Hence, all the transitions from the two- to the one-electron states give the particle leakage current $I$ as

$$I = \sum_{\sigma = \uparrow,\downarrow} \frac{\Gamma_{\sigma,T_+}^{\text{ine}} n_{T_+} + \Gamma_{\sigma,T_-}^{\text{ine}} n_{T_-} + \left( \Gamma_{\sigma,T_+}^{\text{ine}} + \Gamma_{\sigma,T_-}^{\text{ine}} \right) n_{T_+} \Gamma_{\sigma,S_+} n_{S_+} + \Gamma_{\sigma,S_-} n_{S_-}}{2}. \quad (11)$$

Experimentally, the high-bias limit and the zero energy detuning of the levels can be adjusted independently.

Below, the non-linear system of rate equations (9,10) is solved in the stationary limit by analytical and numerical means leading to the leakage current $I$ in Eq. (11).

C. The transition rates

Now we give the rates used in the explicit calculations.

1. The inelastic escape rates

For simplicity, we take the inelastic escape rates to be equal and constant following Refs. [33],[34], i.e.

$$\Gamma_{\sigma,T_+}^{\text{ine}} = \Gamma_{\sigma,T_-}^{\text{ine}} = \Gamma_{\sigma,T_\pm}^{\text{ine}} \equiv \Gamma_{\sigma,T}^{\text{ine}} \quad \text{for} \quad \sigma = \uparrow,\downarrow. \quad (12)$$

Since we consider the limit where the largest of the HF rates dominates over the inelastic escape rates close to the level crossings, then leaving out their energy dependence plays less of a role. Experimentally, the cotunneling rates can be decreased by tuning the energy levels compared to the Fermi levels of the leads (12) while the spin-orbit strength e.g. can be decreased by material choice. Appendix B discusses non-equal inelastic rates.
2. The tunneling rates

The tunneling rates are found by Fermi golden rule. In general, the rates for tunneling into (out of) the DQD are proportional to (one minus) the Fermi distribution of the corresponding lead. Due to the high bias limit, we can leave out the Fermi functions from the explicit expressions of the tunneling rates below.

The non-zero rates for tunneling into the DQD are

\[ \Gamma_{\text{in},\uparrow} = \Gamma_{\text{in},\downarrow} = \Gamma_L \]
\[ \Gamma_{\text{in},\uparrow} = \Gamma_{\text{in},\downarrow} = \frac{\Gamma_L}{2} N \]
\[ \Gamma_{\text{out},\uparrow} = \Gamma_{\text{out},\downarrow} = \frac{\Gamma_L}{2} \left(1 - \frac{p}{N}\right)^2 \]
\[ \Gamma_{\text{out},\uparrow} = \Gamma_{\text{out},\downarrow} = \frac{\Gamma_L}{2} \left(1 + \frac{p}{N}\right)^2 \]

and the non-zero rates for tunneling out of the DQD are

\[ \Gamma_{\text{out},\sigma} = \frac{\Gamma_R p^2}{N^2} \]
\[ \Gamma_{\text{out},\sigma} = \frac{\Gamma_R}{2} \frac{1}{N^2} \text{ for } \sigma = \uparrow, \downarrow \]

i.e. the probability of leaving behind a single electron in the right dot with spin up or down are equal. The rate of leaving T_p goes to zero for p → 0 (i.e. P → 0 or A_L → A_R), since the triplet-singlet mixing vanishes so T_p becomes a blocking state. Here we use \( \Gamma_\alpha = 2\pi |t_\alpha|^2 D_\alpha \) in the standard wide-band approximation where \( D_\alpha \) is the density of states of lead \( \alpha \).

For the calculations to follow, it is helpful to note that they are invariant under the simultaneous interchange of \( \uparrow \leftrightarrow \downarrow, T_+ \leftrightarrow T_- \) and \( S_+ \leftrightarrow S_- \).

3. The hyperfine-induced spin-flip rates

The HF rates are found perturbatively in \( H_{HF} \) using the Fermi golden rule. The HF transition from, say, \( T_+ \) to \( S_+ \) implies a nuclear spin-flip from down to up, so the presence of a spin down among the nuclei is required. Thus, the rate \( W_{S_+,T_+} \) is proportional to the probability of finding a random spin up among the nuclei to be down: \( W_{S_+,T_+} \propto N_s/N \), where \( N_s \) is the number of nuclei with spin \( \sigma = \downarrow \) and \( N = N_s + N_\uparrow \). Likewise the other HF rates \( W_{f,i} \) are proportional to either \( N_\uparrow/N = \frac{1}{2} - P \) or \( N_\downarrow/N = \frac{1}{2} + P \) depending on the direction of the nuclear spin-flip in the process, see Fig. 8.

Here we allow the HF transitions to exchange energy with the environment e.g. by phonons. Phonon emission has been shown to be significant even in low temperature transport experiments. In the transition, it is much easier to emit energy compared to absorbing energy by phonons since the rate for absorbing an energy of \( \hbar \omega \) is proportional to the phonon occupation factor \( n_B(\hbar \omega) \), while the rate for emitting an energy of \( \hbar \omega = n_B(\hbar \omega) + 1 \). Here \( n_B(E) = [e^{E/k_B T} - 1]^{-1} \) is the Bose function. Thus, the absorption rate is suppressed by \( n_B(\hbar \omega)/(n_B(\hbar \omega) + 1) = e^{-\hbar \omega/k_B T} \) compared to the emission rate, which we show below to be crucial for the DNP bistability at zero detuning. The asymmetry between emitting and absorbing energy is not unique to phonons and can also appear from other ways of exchanging energy with a bath due to detailed balance.

Therefore, the two main physical ingredients in the HF rates \( W_{f,i} \) are: (i) the asymmetry between absorbing and emitting energy and, (ii) including the number of the relevant nuclei spin species needed for the transition. A detailed derivation of the rates used here is given in Ref. 12. The non-zero HF rates between the triplets are

\[ W_{T_+,T_+} = W_{T_-,T_-} \]
\[ W_{T_+,T_-} = W_{T_-,T_+} \]

where the phonon part of the rate \( F_{\text{ph}}(E_i - E_f) \) only depends on the difference between the initial and final energies. Note that \( E_{T_+} - E_{T_-} = E_{S_+} - E_{S_-} \) follows from Eq. 8. The non-zero singlet-triplet HF rates are

\[ W_{S_+,T_+} = W_{T_-,S_-} \]
\[ W_{S_+,T_-} = W_{T_-,S_+} \]

where \( E_{T_+} - E_{S_+} = E_{T_-} - E_{S_-} \), so the difference between initial and final energies is the same for e.g. \( W_{S_+,T_+} \) and \( W_{T_-,S_-} \). In the explicit calculations, we use the function

\[ F_{\text{ph}}(E_i - E_f) = \frac{\gamma_{\text{ph}}}{\gamma_{\text{ph}} + (E_i - E_f)^2} c(E_i - E_f) \]

where \( \gamma_{\text{ph}} \) is a characteristic phonon energy scale (e.g. \( \sim 1 \mu \text{eV} \)) and \( c(E_i - E_f) = \theta(E_i - E_f) + \theta(E_f - E_i)e^{-(E_f-E_i)/k_B T} \) is the crucial factor that exponentially
suppresses absorbing compared to emitting energy. Here \( \theta(E) \) is the unit step function. For simplicity, we disregard many details of the phonon description and only include two important aspects: (i) the asymmetry between absorbing and emitting energy and (ii) the further apart the energy levels are, the less probable a transition is — included phenomenologically in the Lorentzian. Moreover, this form includes the limit of HF spin-flips without energy exchange. To get a more detailed phonon description in the rates, both \( H_{\text{HF}} \) and the electron-phonon interaction could be included as perturbations in a \( T \)-matrix approach which gives a description depending on more parameters e.g. the material.

An important difference between the triplet-triplet and singlet-triplet rates, is that the triplet-triplet rates (16) are \( \propto A_2^2 \), whereas the singlet-triplet rates (17) have a common prefactor of \( A_2^2 \) (remembering that \( \nu \propto A_- \)). Thus, the strength of the two kinds of rates near their respective crossings are very different, and the singlet-triplet rates are sensitive to the difference in the Overhauser field between the dots — in contrast to the triplet-triplet rates. However, if \( A_T = A_L \), then all three triplets block transport, since \( \nu = 0 \) so \( \Gamma_{\text{ph}} = 0 \), see Eq. (14a).

Finally, we observe that the HF rates Eqs. (16-17) are invariant under the interchange of

\[
\uparrow \leftrightarrow \downarrow, \quad T_+ \leftrightarrow T_-, \quad S_+ \leftrightarrow S_- \quad \text{and initial} \leftrightarrow \text{final.} \quad (19)
\]

as illustrated in Fig. 3. Similarly, Eq. (10) simplifies to

\[
\dot{P} = \frac{2}{N} \left[ (W_{T_+T_+} - W_{T_-T_-}) n_{T_+} + W_{T_+T_-} n_{T_+} - W_{T_-T_+} n_{T_-} \right]. \quad (21)
\]

In this approximation, the rate equations for the triplet and singlet occupations only couple indirectly through the one-electron occupations. Moreover, we avoid very low temperatures, where the approximation could fail.

To find the stationary DNP, we solve the system of rate equations \((20)-(23)\), i.e. \( \dot{P} = 0 \) and \( n_{\nu} = 0 \) for all \( \nu \). To this end, we subtract \( \dot{n}_i \) Eq. (20g) from \( \dot{n}_i \) Eq. (20f) and use the symmetries under interchange of indices for the tunneling rates (see Eqs. (13)-(15)), i.e. \( \dot{n}_i - \dot{n}_i = (\Gamma_{T_,+} + \Gamma_{S_+,+} + \Gamma_{T_+,p} + \Gamma_{T_+,p} + 2 \Gamma_{\text{ine}})(n_i - n_i) = 0 \), so

\[
\dot{n}_i = n_i \quad (22)
\]

in the stationary limit. Using the symmetries for the tunneling rates and the HF rates, the relation \( (22) \) leads to \( \dot{n}_{T_+} - \dot{n}_{T_-} + \frac{N}{2} \dot{P} = 2 \Gamma_{\text{ine}} (n_{T_+} - n_{T_-}) = 0 \), i.e.

\[
\dot{n}_{T_+} = n_{T_+} \quad (23)
\]

in the stationary limit. Hence, Eq. (21) becomes \( \dot{P} = \frac{N}{2} (W_{T_+T_+} - W_{T_-T_-}) (n_{T_+} + n_{T_-}) = 0 \) using that \( W_{T_+T_-} = W_{T_+T_+} \) and \( W_{T_-T_+} = W_{T_-T_-} \), see Eq. (16).

### III. THE CROSSING OF THE TRIPLETs

Next, we analyze the DNP and leakage current close to the crossing of the three triplet levels near \( B = 0 \). Since \( T_p \) Eq. (16) is not a pure triplet state, it allows for leakage current. As we shall see below, analytical insights — such as the transition temperature — can be achieved from the rate equations (9)-(10) in this case.

#### A. The implicit equation for the nuclear polarization and the simplified rate equations

Now we derive an implicit equation for the DNP from a simplified system of rate equations — valid close to the crossing of the triplet levels. We consider the limit of large energy separation between the triplet and singlet levels compared to \( \gamma_{\text{ph}} \). This can be obtained by large inter-dot tunneling \( \nu \gg \gamma_{\text{ph}} \) and sweeping the magnetic field close to zero. In this limit, we can neglect the singlet-triplet HF rates (17) compared to the triplet-triplet HF rates (16), so the system of rate equations (A1) for equal inelastic escape rates \( \Gamma_{\text{ine}} \), Eq. (12) simplifies to

\[
\begin{align}
\dot{n}_{T_+} &= W_{T_+T_+} n_{T_+} + \Gamma_{T_+,+} n_{T_+} - [W_{T_+T_+} + 2 \Gamma_{\text{ine}}] n_{T_-}, \\
\dot{n}_{T_-} &= W_{T_-T_+} n_{T_+} + \Gamma_{T_-,+} n_{T_+} - [W_{T_-T_+} + 2 \Gamma_{\text{ine}}] n_{T_-}, \\
\dot{n}_{T_p} &= W_{T_pT_+} n_{T_+} + W_{T_pT_-} n_{T_-} + \Gamma_{T_p,+} n_{T_+} + \Gamma_{T_p,+} n_{T_-} - [W_{T_pT_+} + W_{T_-T_+} + \Gamma_{T_p,+} + \Gamma_{T_p,+} + 2 \Gamma_{\text{ine}}] n_{T_p}, \\
\dot{n}_{S_+} &= \Gamma_{S_+,+} n_{T_+} + \Gamma_{S_+,+} n_{T_+} - [\Gamma_{S_+,+} + \Gamma_{S_+,+}] n_{S_+}, \\
\dot{n}_{S_-} &= \Gamma_{S_-,+} n_{T_+} + \Gamma_{S_-,+} n_{T_+} - [\Gamma_{S_-,+} + \Gamma_{S_-,+}] n_{S_-}, \\
\dot{n}_{T_+} &= \Gamma_{S_+,+} n_{S_+} + \Gamma_{S_+,+} n_{S_-} + (\Gamma_{T_p,+} + \Gamma_{S_+,+}) n_{T_+} + \Gamma_{\text{ine}}(n_{T_+} + n_{T_-}) - [\Gamma_{S_+,+} + \Gamma_{S_+,+} + \Gamma_{T_p,+} + \Gamma_{T_p,+}] n_{T_+}, \\
\dot{n}_{T_-} &= \Gamma_{S_-,+} n_{S_+} + \Gamma_{S_-,+} n_{S_-} + (\Gamma_{T_p,+} + \Gamma_{S_-,+}) n_{T_-} + \Gamma_{\text{ine}}(n_{T_+} + n_{T_-}) - [\Gamma_{S_-,+} + \Gamma_{S_-,+} + \Gamma_{T_p,+} + \Gamma_{T_p,+}] n_{T_-}. 
\end{align}
\]

This is not the same as (15) for the tunneling rates, since here the final and initial states are also interchanged.
Important rates close to the crossing of the triplets

\[ \Gamma_{T^+, \sigma} \quad |S_+\rangle \quad \Gamma_{T^-, \sigma} \quad |S_\angle \rangle \quad \Gamma_{T^+, \sigma} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

\[ \Gamma_{T^+, \sigma} \quad W_{T^+, T^\pm} \quad |T^\pm\rangle \quad \Gamma_{T^-, T^-} \quad |T^-\rangle \quad \Gamma_{T^-, T^-} = 0 \]

FIG. 4. (Color online) Illustration of the processes in the rate equations (20) relevant for large energy separation between singlets and triplets and close to the crossing of the three triplets. In this case, only HF rates between the triplets are effective: \( W_{T^+_p, T^\pm} = W_{T^+_p, T^\pm} \) (vertical yellow dashed arrows) polarizing the nuclei negatively (\( dP < 0 \)) and \( W_{T^+_p, T^-} = W_{T^- p, T^-} \) Eq. (16b) (vertical green full arrows) polarizing the nuclei positively (\( dP > 0 \)). The tunneling in and out of the DQD \( \Gamma_{T, s} \) (blue arrows) and the inelastic escape rates \( \Gamma_{\text{ine}} \) (red arrows) are the same as in Fig. 3. In the present case, we show that the stationary DNP is determined by the condition that all triplet-triplet rates are equal, see Eq. (24).

Thus, since the occupations are positive, we arrive at

\[ W_{T^+_p, T^\pm} = W_{T^+_p, T^\pm}, \quad (24) \]

which is the implicit equation for the steady state DNP \( P \) — remembering that both rates depend on \( P \) explicitly and through the Overhauser split energy levels \( E_{T^\pm} \), see Eq. (16). Equivalently, this can be written as \( W_{T^+_p, T^\pm} = W_{T^- p, T^-} \). Physically, the relation (24) means that the DNP stabilizes at a value such that the phonon emission and absorption transitions between the two levels \( T^\pm \) and \( T^- \) become equally probable. Inserting Eq. (16), the relation (24) can be rewritten as

\[ 1 + P = \frac{\mathcal{F}_{ph}(E_{T^\pm} - E_{T^-})}{\mathcal{F}_{ph}(E_{T^-} - E_{T^-})}. \quad (25) \]

This shows that the DNP is insensitive to the part of \( \mathcal{F}_{ph} \), which is even in energy, and only depends on the difference between emitting and absorbing energy. In other words, the even-energy part of the function \( \mathcal{F}_{ph} \) cancels out on the right-hand side of Eq. (25) and we are left with the ratio between absorbing and emitting energy in the transition. Hence, the DNP is largely independent of the way the phonons are modelled in the HF rates, as long as the important asymmetry between emitting and absorbing energy is included.\(^{[129]}\) We observe a crucial difference to previous works\(^{[130,129]}\) where non-zero DNP is induced only at finite detuning, since emitting and absorbing energy is modelled as being equally likely in these works, see Sec. I A. By inserting the energies Eq. (8) into Eq. (25), we end up with the following implicit equation for \( P \),

\[ P = \frac{\tan(g \mu_B B + \frac{1}{2} A_{c,t} P)}{2 k_B T}. \quad (26) \]

From this equation, it is not possible to obtain a closed analytical expression for \( P \). Remarkably, it shows that \( P \) near the crossing of the triplet levels only depends on the Zeeman splitting \( g \mu_B B / A_+ \) and the temperature \( k_B T / A_+ \) — both measured in units of \( A_+ = (A_L + A_R) / 2 \).

The form of the implicit equation (26) resembles the one found by using mean-field theory to a Heisenberg spin model, which describes an equilibrium ferromagnetic phase transition driven by temperature\(^{[112]}\). In contrast, here the polarization is dynamically induced by the leakage current, i.e. a non-equilibrium situation.

The DNP versus \( T \) and \( B \) is easily found numerically from Eq. (26), see Fig. 3. For \( B = 0 \), the DNP has a bifurcation at a certain transition temperature \( T_{c,tt} \), where the system goes from one stable DNP for \( T > T_{c,tt} \) to two stable DNPs and an unstable one for \( T < T_{c,tt} \). The stability of the DNP is found by numerical iteration of the set of differential equations\(^{[125]}\). The transition temperature for \( B = 0 \) is readily found from Eq. (26) to be

\[ k_B T_{c,tt} = \frac{1}{4} A_+. \quad (27) \]

This is a remarkably simple and insightful result. It shows that the transition temperature \( T_{c,tt} \) is given only by the average HF constants \( A_+ = (A_L + A_R) / 2 \). For \( T < T_{c,tt} \), the DNP can have two stable values and therefore so can the current. Hence, the current shows...
First, we find the stationary occupations using \( n_\downarrow = n_\uparrow \) (Eq. (22)), \( n_{T_\uparrow} = n_{T_\downarrow} \) (Eq. (23)) and the implicit equation (24) for \( P \). Now, subtracting \( \dot{n}_{S_\downarrow} \) (20a) and \( \dot{n}_{S_\uparrow} \) (20c) using \( n_\downarrow = n_\uparrow \) and the index invariances of \( \Gamma_i \), Eqs. (13-15), we find \( \dot{n}_{S_\downarrow} - \dot{n}_{S_\uparrow} = 2\Gamma_\uparrow \downarrow (n_{S_\downarrow} - n_{S_\uparrow}) = 0 \), so

\[
n_{S_\downarrow} = n_{S_\uparrow}
\]

in the steady state. This simplifies the sum of \( \dot{n}_{S_\uparrow} \) (20a) and \( \dot{n}_{S_\downarrow} \) (20c), \( \dot{n}_{S_\downarrow} + \dot{n}_{S_\uparrow} = 0 \), and leads to

\[
n_{S_\downarrow} = \frac{\Gamma_{S_\downarrow \uparrow} + \Gamma_{S_\uparrow \downarrow}}{2\Gamma_\uparrow \downarrow} n_\uparrow
\]

by again using \( n_\downarrow = n_\uparrow \) and the index exchange symmetries of \( \Gamma_i \), Eqs. (13-15). Inserting these occupation relations into the normalization condition \( \sum_n n_n = 1 \), we get

\[
n_\downarrow = \frac{\Gamma_{S_\downarrow \uparrow}}{\Upsilon} [1 - n_{T_\downarrow} - 2n_{T_\uparrow}],
\]

where \( \Upsilon \equiv 2\Gamma_{T_\downarrow \uparrow} + \Gamma_{S_\downarrow \uparrow} + \Gamma_{S_\uparrow \downarrow} \). By inserting Eqs. (22), (23), (24), (25), (20) and (30) into \( n_{T_\downarrow} = 0 \) (20c) and \( n_{T_\uparrow} + n_{T_\downarrow} = 0 \) (20b), we obtain two coupled equations for the occupations \( n_{T_\downarrow} \) and \( n_{T_\uparrow} \) with the solution

\[
n_{T_\downarrow} = \frac{\Gamma_{T_\downarrow \uparrow}}{\Lambda} \left( \Gamma_{S_\downarrow \uparrow} W_{T_\downarrow, T_\uparrow} (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{T_\uparrow \downarrow}) \right),
\]

\[
n_{T_\uparrow} = \frac{\Gamma_{T_\downarrow \uparrow}}{\Lambda} \left( \Gamma_{T_\downarrow \uparrow} W_{T_\downarrow, T_\uparrow} (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{T_\uparrow \downarrow}) \right)
\]

where

\[
\Lambda = \left( \frac{2\Gamma_{T_\downarrow \uparrow} (2\Gamma_{T_\downarrow \uparrow} + 3\Gamma_{T_\downarrow \uparrow}) + 3\Gamma_{T_\downarrow \uparrow}}{2\Gamma_{T_\downarrow \uparrow} (2\Gamma_{T_\downarrow \uparrow} + 3\Gamma_{T_\downarrow \uparrow})} \right)
\]

The explicit expressions for \( n_\downarrow = n_\uparrow \) can easily be found by inserting Eq. (31a) into Eq. (30). This in turn leads to the expression for \( n_{S_\downarrow} = n_{S_\uparrow} \) via Eq. (29). Thus, we now have all the stationary occupations close to the crossing of the triplets in terms of the rates.

The leakage current in the high bias limit is now obtained by inserting the occupations into Eq. (11), i.e.

\[
I = \frac{2\Gamma_{T_\downarrow \uparrow}}{\zeta} \left( \Gamma_{T_\downarrow \uparrow} (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{S_\downarrow \uparrow} + \Gamma_{S_\uparrow \downarrow} + \Gamma_{T_\uparrow \downarrow}) \right)
\]

where we introduced

\[
\zeta \equiv (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{T_\downarrow \uparrow}) (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{S_\downarrow \uparrow} + \Gamma_{T_\uparrow \downarrow} + \Gamma_{T_\uparrow \downarrow}) + 2\Gamma_{T_\downarrow \uparrow} \Gamma_{T_\downarrow \uparrow} W_{T_\downarrow, T_\uparrow} (2\Gamma_{T_\downarrow \uparrow} + \Gamma_{T_\uparrow \downarrow}) \]

1. Analytical stationary occupation and current expressions in terms of the rates

Next, we find the leakage current close to the crossing of the triplets using the simplified rate equations (20-21).
Thus, the current is expressed in terms of the dimensionless triplet-singlet mixing parameter \( p \) in Eq. (31) and the three rates, \( \gamma_{RL} \), \( \gamma_{ine} \) and \( w \) — all measured in units of the basic tunneling rate \( \Gamma_L \). The asymmetry between the coupling of the DQP to the left and right lead is described by \( \gamma_{RL} \), where \( \gamma_{RL} = 1 \) for the symmetric case. Hence, \( \gamma_{RL} \) is on the order of unity. In contrast, both the dimensionless HF triplet-triplet rates \( \Gamma^{tt}_L \) and \( \Gamma^{tt}_R \) and the dimensionless inelastic escape rate \( \gamma_{ine} \) are much smaller than unity: \( \gamma_{ine}, w \ll \gamma_{RL} \sim 1 \). Moreover, here we focus on the limit of the inelastic escape rate being much smaller than the HF rate close to the crossing of the triplets.

We note that without singlet-triplet mixing, \( p = 0 \) (i.e. \( P = 0 \) or \( A_L = A_R \)), the current (33) reduces to

\[
\frac{I(p=0)}{\Gamma_L} = \frac{8\gamma_{RL}\gamma_{ine}}{3\gamma_{RL} + 2\gamma_{ine}(1 + 2\gamma_{RL})},
\]

which is independent of the HF rate \( w \). Physically, this can be understood in the following way. For \( p = 0 \), the escape channel from \( T_p \) due to singlet-triplet mixing disappears. Thus, for \( p = 0 \) only the inelastic escape channel \( \Gamma^{ine} \) contributes to the current through the three triplet states (remembering that the singlet levels are far away in energy). Since we use equal inelastic escape rates \( \Gamma^{ine} \) from the three triplet states here, then the current does not depend on from which triplet state the electron tunnels out. Thus, the current has to be independent of the HF transitions between the three triplets for \( p = 0 \) as found in Eq. (33). In contrast, if the inelastic escape rates from the three triplets are not equal, then the current can indeed depend on the HF rate even for \( p = 0 \). An example of this, is given in Eq. (B3) in Appendix B.

We stress that the rates still depend on the DNP, which in general is not analytically known in terms of the external parameters. Thus, the occupations (31) and current expressions (32-33) are also not explicit functions of the external parameters. To obtain explicit expression versus external parameters, the DNP needs to be found from the implicit DNP equation (26). This will be done below.

2. The leakage current versus magnetic field: Emergence of hysteresis below the transition temperature

Next, we analyze the leakage current versus \( B \)-field as shown in Fig. 7 for \( T < T_{c,TT} \), \( T = T_{c,TT} \) and \( T > T_{c,TT} \). To this end, the DNP \( P \) is found numerically from the implicit equation (26) [Fig. 7(a,d,g)] and then inserted into the current expressions (33) [Fig. 7(b,e,h)]. Hence, if multiple stable DNP solutions exist, then there will also be multiple possible stable values of the current. The actually stationary leakage current and DNP in a concrete situation therefore depend on the initial value in time of the DNP as in other non-linear dynamical systems.

The hysteresis in the current comes about in the following way: Consider \( T < T_{c,TT} \) and the magnetic field tuned so high that there is only a single DNP solution, e.g. \( g\mu_B B = 7 \mu\text{eV} \) in Fig. 7(a). By decreasing \( g\mu_B B \) one will enter the region of multiple possible DNP’s [at \( g\mu_B B \sim 2.34 \mu\text{eV} \) in Fig. 7(a)]. Since the DNP is a stable solution against small fluctuations, the system will remain on the upper stable branch (\( P > 0 \)) until the critical \( B \)-field, where the upper branch cease to exist [about \( g\mu_B B \sim -2.34 \mu\text{eV} \) in Fig. 7(a)]. At this critical field, the system has to go to the lower stable DNP branch (with \( P < 0 \)). Thus, the DNP change discontinuously versus \( B \). This in turn leads to a jump in the current (as seen in the blue full curve on Fig. 7(b)) for sweeping the field backwards from a high value). For \( B \)-fields lower than the critical one, the DNP is single valued again and so is the current. Now, if at this point the field is increased beyond the critical field [at \( g\mu_B B \sim -2.34 \mu\text{eV} \) in Fig. 7(a)] one will follow the lower stable DNP branch with \( P < 0 \), leading to the dashed red curve in Fig. 7(b). This sweep direction also leads to a sharp jump once the lower stable DNP branch cease to exist [at \( g\mu_B B \sim +2.34 \mu\text{eV} \)]. Thus, the hysteretic behavior of the leakage current for \( T < T_{c,TT} \) is now evident. The discontinuity versus \( g\mu_B B \) for \( T > T_{c,TT} \) is also seen in the energy levels Fig. 7(c), where only one sweeping direction is shown for clarity.

We observe that the width of the hysteresis loop increases with decreasing \( T \), since this width is given by the vertical distance between the two (full) lines in Fig. 6.
Identifying that the transition temperature $T_{c,\text{TT}}$ simply is given by the average HF constants (27), is an important result of this paper. Experimentally, the HF constants are of order $100\mu$eV, so $T_{c,\text{TT}}$ is on the order of 0.3K, which is within range of modern experiments.

To test the results of the simplified model without HF triplet-singlet rates presented in Fig. 7, we have numerically iterated the full set of rate equations (A1,10) including all rates. For the parameters of Fig. 7, where the $E_{Sz}$ levels are far away from the triplet levels – the two calculations give the same results (not shown in the figure), i.e. the presented simplified model works well.

Now we give a better understanding of the form of the current versus $B$-field. In this work, we focus on the limit where the HF rates dominate the inelastic escape rate close to the level crossings as in Refs. 35,36. Nevertheless, the inelastic escape rate plays an important role for the current in the following. First, we analyze in detail current versus $g\mu_B B$ for $T \geq T_{c,\text{TT}}$, where the DNP is single valued and therefore no current hysteresis is found [Fig. 7(e,h)]. The DNP goes continuously through $P = 0$ at $g\mu_B B = 0$. As discussed above (see Eq. (34)), the singlet-triplet mixing disappears at $P = 0$, which in turn closes the escape path from $T_p$ as $\Gamma_R \Gamma_L \sim P^2$ for $|P| \ll 1$, see Eq. (14a). Thus, the current decreases for $B \rightarrow 0$ and $T \geq T_{c,\text{TT}}$ to a value only given by the inelastic escape rate – even though it is weak. For $\gamma_R = 1$ Eq. (34) gives

$$\frac{I(B = 0, T \geq T_{c,\text{TT}})}{\Gamma_L} = \frac{8\gamma_{\text{ine}}}{3 + 6\gamma_{\text{ine}}},$$

which agrees perfectly with the value of $2.7 \times 10^{-6}$ found in Fig. 7(e,h) for $B = 0$. The slope of $P$ at $g\mu_B B = 0$ increases rapidly when approaching $T = T_{c,\text{TT}}$ from temperatures above $T_{c,\text{TT}}$, see Fig. 7(d,g). Hence, the dip in the current at $g\mu_B B = 0$ becomes increasingly sharper.
Hyperfine and Inelastic rates for $T > T_{c,TT}$

![Zoom on rate crossing](image)

FIG. 8. (Color online) The dimensionless HF rate $w$ (black full line) and inelastic escape rate $\gamma_{ine} = 1/\Gamma_L$ (gray dashed line) versus magnetic field $g\mu_B B$ for $T = 1.2T_{c,TT}$. The inset shows that $\gamma_{ine} \gg w$ for $|g\mu_B B| \gtrsim 2.7\mu eV$. In contrast, the HF rate $w$ dominates by orders of magnitude over $\gamma_{ine}$ around $B = 0$. The parameters are the same as in Fig. 7(g-i).

for $T$ approaching $T_{c,TT}$, see Fig. 7(e,h) and the inset.

The HF rates $w$ goes to zero as the triplet energies move apart, i.e. $w \to 0$ for increasing $|g\mu_B B|$, see Fig. 8. Thus, the inelastic escape rate $\gamma_{ine}$ will eventually become larger than $w$, since $\gamma_{ine}$ is constant. For the values used in Fig. 7(g-i), we have $\gamma_{ine} > w$ for $|g\mu_B B| \gtrsim 2.7\mu eV$ as shown in the inset of Fig. 8. Thus, once the triplet levels move further apart, the triplet-triplet rate $w$ goes to zero and can be neglected in the current (33). That corresponds to $|g\mu_B B| \gtrsim 2.7\mu eV$ for $T > T_{c,TT}$ in the numerical example of Fig. 7(g-i). For $\gamma_{RL} = 1$, we find (32)

$$
\frac{I_{w \to 0}}{I_L} = \frac{8\gamma_{ine}[\gamma_{ine} + (1 + \gamma_{ine})p^2]}{2p^2 + \gamma_{ine}(3 + 2p^2)[1 + 2\gamma_{ine} + 2(1 + \gamma_{ine})p^2]} = 4\gamma_{ine} + O[(\gamma_{ine})^2].
$$

(36)

Here $\gamma_{ine} \ll |p|$ for $|P| > 0.1$ for $A_-$ and $t$ similar to those used in Fig. 7. In Figs. 7(b,e,h), we see that the current levels off to a constant value of $\sim 4 \times 10^{-6}$ far away from the level crossing, which is in perfect agreement with the prediction (36). Therefore, we have found that

$$
\frac{I(B = 0, T \geq T_{c,TT})}{I_{w \to 0}} \approx \frac{2}{3} \quad \text{for} \quad \gamma_{ine} \ll 1,
$$

(37)

so the value at the crossing for $B = 0$ (for $T \geq T_{c,TT}$) is generically lower than the value that the current levels off to asymptotically [Fig. 7(e,h)].

Now we have shown that the current value both at the crossing and far away form the crossing of the triplets is determined by the inelastic escape rate. Albeit the HFI of course is essential in having a DNP in the first place. Next, we discuss how the HF triplet-triplet transitions can increase the leakage current close to $-\Delta$ but not exactly at $-\Delta$ the level crossing. At the crossing ($B = 0$ for $T \geq T_{c,TT}$), all three triplet states form the bottleneck for transport through the DQD. For $P \neq 0$, the triplet-singlet mixing leads to the additional escape from $T_p$ such that the transport bottleneck (far away from the crossing) now only consist of $T_\pm$. For $T \geq T_{c,TT}$, we only get $P \neq 0$ for $B \neq 0$. Moreover, note that $n_{T_p}$ becomes negligible compared to $n_{T_+}$ far from the level crossing for $T \geq T_{c,TT}$ (see Fig. 9). The point is that in the region close to the level crossing, the HF triplet-triplet transitions leads to an escape path from $T_\pm$ via $T_p$. Since the HF rate $w$ is much larger than $\gamma_{ine}$ close to the crossing, this escape route is so effective that it creates the side-peaks of the current at finite $|g\mu_B B|$ seen in Fig. 7(e,h). These current side-peaks are therefore sensitive to the value of the inelastic rate: if $\gamma_{ine}$ is increased by a factor of 5 or more, then the side-peaks in Fig. 7(h) disappear. In contrast, if $\gamma_{ine}$ is decreased the side-peaks remain. Fig. 9 shows that near the current side-peaks [with maxima at $g\mu_B B \approx \pm 1.4\mu eV$ in Fig. 7(h)], the occupations $n_{T_\pm}$ are much larger than $n_{T_p}$.

Finally, we point out that the form of the current for $T < T_{c,TT}$ can be understood by using the above considerations, but taking into account that the DNP jumps between rather high values [e.g. $P \approx 0.45$ to $P \approx -0.8$ at $g\mu_B B \approx 2.3\mu eV$ in Fig. 2(a)]. Therefore, the rich region around $P = 0$ is simply skipped. [Note also the vertical scale change in Fig. 2(b) compared to (e) and (h).]

Therefore, we have now obtained an understanding of the leakage current versus magnetic field close to the crossing of the triplet levels under the assumption that the singlet levels $E_{S\pm}$ are far away in energy.

3. The current in the high temperature and low $B$-field limit

Next, we show that the leakage current can be given analytically in terms of the external parameters for low magnetic fields and $T > T_{c,TT}$. Specifically, if
Important processes close to the singlet-triplet crossing

$$\frac{2g\mu_B B}{A_L + P} \ll T/T_{c,TT}$$ then the hyperbolic tangent in the implicit DNP equation \([20]\) can be expanded, so

$$P \approx \frac{1}{2} g\mu_B B/k_B \left( T - T_{c,TT} \right).$$

(38)

This is similar to a Curie-Weiss law for a ferromagnet in the paramagnetic region and the fact that \(P \propto (T - T_{c,TT})^{-1}\) is typical for the mean-field approach used here.\([20]\) Expanding the current \([33]\) in \(P\) and inserting Eq.\([38]\), the current for low \(B\)-fields explicitly becomes:

$$I(T > T_{c,TT}) \approx \frac{8 \gamma_{RL} \gamma_{line}}{3 \gamma_{RL} + 2 \gamma_{line} (1 + 2 \gamma_{RL})} \left( g\mu_B B \right)^2 \left[ k_B (T - T_{c,TT}) \right].$$

(39)

This describes the current dip close to \(B = 0\) seen in Fig.\(7\)h. It shows explicitly that the current increases by changing slightly \(B\) away from \(0\), since \(\gamma_{line} < 1 + \sqrt{17} \gamma_{RL}/8\) for reasonable parameters. We observe that the HF rate \(w\) only appears beyond the second order term, however, already this term contains \(A_L = (A_T - \gamma_{RL}/2)\). The lowest order term for \(B = 0\) coincides with Eq.\([31]\) for no triplet-singlet mixing as expected.

IV. THE SINGLET-TRIPLET CROSSING

In this section, we analyse the DNP and leakage current close to the crossing of the singlet levels \(E_{S_L}\) and the pure triplet levels \(E_{T_+}\) at finite magnetic field. Since we consider zero detuning, the crossing of the levels always happens in pairs, e.g. \(E_{T_-}\) and \(E_{S_F}\) cross at the same \(B\)-field as \(E_{T_+}\) and \(E_{S_L}\) do. Interestingly, here we find that the transition temperature \(T_{c,ST}\) for the singlet-triplet level crossing is enhanced compared to \(T_{c,TT}\) Eq.\([27]\).

A. A simplified model for the singlet-triplet crossing and its implicit polarization equation

Next, we develop a simplified set of rate equations valid close to the crossing of \(E_{T_+}\) \((E_{T_-})\) and \(E_{S_L}\) \((E_{S_F})\) for positive \(B\)-field splitting, \(g\mu_B B > 0\). From these equations, we derive an implicit equation for the DNP. The level crossings for \(g\mu_B B < 0\) follow along similar lines.\([13]\)

As a first approach, one might intend to follow the same strategy as in Sec.\([IIA]\) for the crossing of the triplets: Keep only the HF rates between the levels, which are close in energy. Hence, we keep only the HF terms in Eqs.\([10]\) involving \(W_{T_\pm, S_F}\) and \(W_{S_F, T_\pm}\) in the present case. Such a simplification leads to \(W_{S_F, T_+} = W_{T_+, S_F}\) as an implicit equation for the DNP — much like in the case of the crossing of the triplets Eq.\([21]\). This leads to the same transition temperature as \(T_{c,TT}\) Eq.\([27]\) to a very good approximation. However, for the singlet-triplet level crossing, this approach is actually not a good approximation. Explicitly, we find that this approach does not reproduce the DNP found by a numerical iteration of the rate equations \([10][11]\) including all rates (for an inter-dot coupling \(t\) about two orders of magnitude larger than \(\gamma_{ph}\))\([13]\). The approach fails for the following reasons: Firstly, the occupations \(n_{S_F}\) are much smaller than the occupations \(n_{T_\pm}\) and \(n_{S_F}\) since escape from the singlets is much easier than from the triplets in the SB regime. Secondly, the triplet-triplet and the singlet-triplet HF rates have the overall prefactors of \(A_T^2 + A_L^2\), respectively, such that the triplet-triplet rate is enhanced compared to the singlet-triplet rate (for comparable energy level splitting). Due to these two facts, the triplet-triplet HF terms can still be comparable in magnitude to the singlet-triplet terms in the rate equations \([10]\) — even though \(|E_{T_+} - E_{T_-}| \gg \gamma_{ph}\) close to the singlet-triplet crossing. In other words, we cannot neglect terms like \(W_{T_+, T_-} n_{T_+}\) compared to terms like \(W_{S_F, T_+} n_{T_+}\).

Here, we have to adopt a different approach of simplifying the rate equations from the one used for the crossing of the triplets in Sec.\([IIA]\). This is done in order to describe the regime of large singlet-triplet energy splitting \(|E_{S_F} - E_{T_+}| \gg \gamma_{ph}\); however, not so large that the triplet-triplet rates cannot still play a role. Our approach is to neglect two kinds of terms in the rate equations \([10]\). (I) The HF singlet-triplet terms between \(T_+\) \((T_-)\) and \(S_+\) \((S_-)\) can safely be neglected, because of
large energy separation (for $g\mu_B B > 0$) combined with an overall prefactor of $A^2$ in the rate. Thus, we neglect terms of the form $W_{T_s,S_+}n_{S_+}$ and $W_{S_-,T_s}n_{T_s}$, where $\nu = \pm$ and $\nu = -\nu$. (ii) Due to the SB regime, $n_{S_\nu} \ll n_{T_s}$, for $\nu', \nu = \pm$, so we neglect the terms $W_{T_\nu,S_\pm}n_{S_\pm}$. We thereby neglect HF transitions from $S_\pm$ to $T_\pm$, since tunneling out from $S_\pm$ are much more probable processes. Using equal inelastic escape rates $[12]$, these two simplifications lead to the following rate equations for $g\mu_B B > 0$

\[
\begin{align*}
\dot{n}_{T_\nu} &= W_{T_\nu, T_s} n_{T_s} + \Gamma_{T_\nu, T_s} n_{T_s} - [W_{S_+, T_\nu} + W_{T_\nu, T_s} + 2\Gamma^\text{ine}] n_{T_\nu}, \\
\dot{n}_{T_\nu} &= W_{T_\nu, T_s} n_{T_s} + \Gamma_{T_\nu, T_s} n_{T_s} - [W_{S_-, T_\nu} + W_{T_\nu, T_s} + 2\Gamma^\text{ine}] n_{T_\nu}, \\
\dot{n}_{T_\nu} &= W_{T_\nu, T_s} n_{T_s} + \Gamma_{T_\nu, T_s} n_{T_s} - [W_{S_+, T_\nu} + W_{T_\nu, T_s} + 2\Gamma^\text{ine}] n_{T_\nu}, \\
\dot{n}_{S_\pm} &= W_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, S_\pm} n_{S_\pm} - [\Gamma_{T_\nu, S_\pm} + \Gamma_{S_\pm, S_\pm}] n_{S_\pm}, \\
\dot{n}_{S_\pm} &= W_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, S_\pm} n_{S_\pm} - [\Gamma_{T_\nu, S_\pm} + \Gamma_{S_\pm, S_\pm}] n_{S_\pm}, \\
\dot{n}_{S_\pm} &= W_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, T_s} n_{T_s} + \Gamma_{S_\pm, S_\pm} n_{S_\pm} - [\Gamma_{T_\nu, S_\pm} + \Gamma_{S_\pm, S_\pm}] n_{S_\pm}, \\
\dot{n}_{S_\nu} &= \Gamma_{S_\nu, S_\nu} n_{S_\nu} - \Gamma_{S_\nu, S_\nu} n_{S_\nu} - [\Gamma_{T_\nu, S_\nu} + \Gamma_{S_\nu, S_\nu}] n_{S_\nu}, \\
\dot{n}_{S_\nu} &= \Gamma_{S_\nu, S_\nu} n_{S_\nu} - \Gamma_{S_\nu, S_\nu} n_{S_\nu} - [\Gamma_{T_\nu, S_\nu} + \Gamma_{S_\nu, S_\nu}] n_{S_\nu}, \\
\dot{n}_{S_\nu} &= \Gamma_{S_\nu, S_\nu} n_{S_\nu} - \Gamma_{S_\nu, S_\nu} n_{S_\nu} - [\Gamma_{T_\nu, S_\nu} + \Gamma_{S_\nu, S_\nu}] n_{S_\nu}.
\end{align*}
\]

Likewise, Eq. (10) for the DNP simplifies to

\[
\dot{P} = \frac{2}{N} \left( (W_{T_\nu, T_s} - W_{T_\nu, T_s}) n_{T_\nu} + (W_{S_\pm, T_s} + W_{T_\nu, T_s}) n_{T_\nu} - (W_{S_-, T_\nu} + W_{T_\nu, T_s}) n_{T_\nu} \right).
\]

The simplified model is illustrated on Fig. 10. The only difference compared to the rate equations (20–21) for the crossing of the triplets, is the direct coupling of the singlets and triplets via the terms $W_{S_\pm, T_s} n_{T_s}$. Now we derive the implicit equation for the stationary DNP from these rate equations. We do not use the explicit form of the rates, but only the invariances under index interchange $[15, 19]$. We begin by noting that $\dot{n}_1 - \dot{n}_1 = 0$ leads to $n_1 = n_1$ in the stationary state. Inserting this into $\dot{n}_1 - \dot{n}_1 + (N/2) \dot{P} = 0$ gives $n_{T_\nu} = n_{T_\nu}$ in the steady state. These two relations are the same as in the description of the triplet level crossing, see Eqs. (22–23). In fact, $n_\pm = n_\nu$ and $n_{T_\pm} = n_{T_\nu}$ can be derived form the rate equations $[11, 10]$ including all rates and equal inelastic rates. However, at this point the two descriptions separate, since the stationary singlet occupations are no longer equal [as in Eq. (28)]. Instead, we find

\[
n_{S_\nu} = \frac{\left( \Gamma_{S_\nu, S_\nu} + \Gamma_{S_\nu, S_\nu} \right) n_{S_\nu} + W_{S_\pm, T_s} n_{T_\nu}}{2\Gamma_{T_\nu, S_\nu}},
\]

by solving $\dot{n}_{S_\nu} = 0$ using $n_{T_\nu} = n_{T_\nu}$ and that $\Gamma_{T_\nu, S_\nu} \neq 0$. Hence, $n_{S_\nu} \neq n_{S_\pm}$ if and only if $W_{S_-, T_\nu} \neq W_{S_+, T_\nu}$. These relations are used to derive

\[
\begin{align*}
n_{T_\nu} &= n_{T_\nu} \left( 2\Gamma_{T_\nu, T_s} (\Gamma^\text{ine} + \Gamma_{T_\nu, T_s}) \right) \\
&\quad + (\Gamma_{T_\nu, T_s} + \Gamma_{T_\nu, T_s}) (W_{T_\nu, T_s} + W_{T_\nu, T_s}), \\
\dot{n}_{T_\nu} &= n_{T_\nu} \left( 2\frac{2}{\Gamma_{T_\nu, T_s}} \left( W_{T_\nu, T_s} + W_{T_\nu, T_s} \right) \right) \\
&\quad + (\Gamma_{T_\nu, T_s} + \Gamma_{T_\nu, T_s}) (W_{T_\nu, T_s} + W_{T_\nu, T_s}).
\end{align*}
\]

from $\dot{n}_1 + \dot{n}_1 = 0$ and $\dot{n}_{T_\nu} = 0$. Here we introduced the non-zero quantity

\[
\kappa \equiv 8(\Gamma^\text{ine})^2 + 2(\Gamma_{T_\nu, T_s} + 3\Gamma^\text{ine}) (W_{T_\nu, T_s} + W_{T_\nu, T_s}) + 8\Gamma^\text{ine} \Gamma_{T_\nu, T_s} + (W_{S_+, T_\nu} + W_{S_-, T_\nu}) \times (W_{T_\nu, T_s} + W_{T_\nu, T_s}).
\]

Note that the triplet occupation expressions are proportional to the one-electron occupation. By inserting the occupation expressions into Eq. (11), we find $\dot{P} = n_{T_\nu} / 2 N$, where $\chi$ is a combination of rates (given below). In order to satisfy $\dot{P} = 0$ in steady state, we have to require that $\chi = 0$, since the occupation is positive. Thus, we arrive at the implicit DNP equation, $\chi = 0$, which explicitly is

\[
0 = (W_{T_\nu, T_s} - W_{T_\nu, T_s}) \left\{ \Gamma_{T_\nu, T_s} (\Gamma_{T_\nu, T_s} + \Gamma^\text{ine}) + (W_{T_\nu, T_s} + W_{T_\nu, T_s}) (\Gamma_{T_\nu, T_s} + \Gamma_{T_\nu, T_s}) + \Gamma_{T_\nu, T_s} \left[ \frac{1}{2} (W_{S_+, T_\nu} + W_{S_-, T_\nu}) + 2\Gamma^\text{ine} \right] \right\} \\
+ (W_{S_-, T_\nu} - W_{S_+, T_\nu}) \left\{ \Gamma_{T_\nu, T_s} (\Gamma_{T_\nu, T_s} + \Gamma^\text{ine}) + \frac{1}{2} (W_{T_\nu, T_s} + W_{T_\nu, T_s}) (\Gamma_{T_\nu, T_s} + \Gamma_{T_\nu, T_s}) \right\} = \chi.
\]
This implicit equation for $P$ is more involved than the one describing the DNP around the crossing of the triplet levels Eq. (24). Moreover, a simple formula for the transition temperature $T_{c,ST}$ is not immediately apparent. Nevertheless, the implicit equation (44) does give some insights. For instance, it describes the crossing of the triplets as a special case: Close to the crossing of the triplets, $W_{ST,TS} \approx$ negligible such that Eq. (44) simplifies to $W_{ST,TS} = 0$, which is exactly Eq. (21).

Furthermore, the implicit equation (44) shows that $T_{c,ST}$ stems from the asymmetry between energy emission and absorption in the HF process. This we show by assuming the opposite: absorbing or emitting an energy in the HF process is equally likely [i.e. $F_{ph}(E)$ is even in Eqs. (16, 17)]. This assumption leads to $W_{S,T} - W_{S,T} \propto P$ and $W_{T,ST} = W_{T,ST} \propto P$ by using Eqs. (16, 17), such that the implicit equation (44) can be written as $0 = PG(P)$, where $G(P)$ is a strictly positive function. Thus, $P = 0$ is the only DNP solution without the asymmetry between emission and absorption of energy such that no DNP bistability occurs.

Here, we find the DNP from the implicit equation (44) numerically. This in turn gives the leakage current and transition temperature $T_{c,ST}$ as we will discuss next.

### B. The nuclear polarization, leakage current and the singlet-triplet crossing transition temperature

We extract the DNP versus magnetic field for various temperatures numerically from the implicit equation (44), see Fig. 11(a,d,g). In this way, we can pinpoint the region of temperature and $B$-field with one and three DNP solutions, respectively, as shown in Fig. 12. This in turn allows to determine the transition temperature $T_{c,ST}$ for the singlet-triplet crossing, where the DNP becomes single-valued. In the specific case of parameters in Fig. 11 and 12 we find $T_{c,ST} \approx 2.80 A_{\gamma}/k_{B}$, which is about one order of magnitude larger than $T_{c,ST} = A_{\gamma}/(4k_{B})$ in Eq. (27).

We have repeated this procedure to find the transition temperatures for different parameters as seen in tables I and II. We find that the transition temperature $T_{c,ST}$ depends on various external parameters — in contrast to the crossing of the triplets, where $T_{c,ST} = A_{\gamma}/(4k_{B})$. In table I, we find that $T_{c,ST}$ seems largely insensitive to decreasing the inelastic rate $\gamma_{ine} = \Gamma_{ine}/\Gamma_{L}$ as long as it is smaller than the dominant singlet-triplet rates close to the level crossing. This makes sense from the implicit equation (44), since a small $\Gamma_{ine}$ is negligible compared to $W_{TS,ST}$ and $\Gamma_{ST,TT}$. Table II also reveals a small non-monotonic dependence of $T_{c,ST}$ on $t$, which controls the level splitting $|E_{ST,TS} - E_{TT}|$, and, in turn, the size of the HF triplet-triplet rates close to the singlet-triplet crossing.

An effective way to change the relative magnitudes of the singlet-triplet and the triplet-triplet rates, is to change $A_{\gamma}$ since the singlet-triplet rate have an overall prefactor of $A_{\gamma}^{2}$ (whereas $W_{TT,ST} \propto A_{\gamma}^{2}$). Table II shows $T_{c,ST}$ for varying the relative strength of the singlet-triplet and triplet-triplet rates. The largest transition temperature, $k_{B}T_{c,ST} \approx 6.94 A_{\gamma}$, is found when the singlet-triplet, triplet-triplet and inelastic rates all are of the same order. In contrast, the smallest value, $k_{B}T_{c,ST} \approx 1.32 A_{\gamma}$, is found when the singlet-triplet rate dominates by more than two orders of magnitude over the triplet-triplet rate. Moreover, the number of nuclei change $T_{c,ST}$ slightly. Finally, we remark that $T_{c,ST}$ is not simply proportional to $A_{\gamma}$. Nevertheless, we give $k_{B}T_{c,ST}$ in units of $A_{\gamma}$ in order to compare it with a typical energy scale of the problem. Altogether, a common feature for all the parameters considered here, is that $T_{c,ST}$ is found to be larger than $T_{c,TT}$.

The leakage current is found from Eq. (11) by inserting the stable DNP found from the implicit equation (44). To this end, we use $\sum_{\nu} n_{\nu} = 1$ to specify all the occupations. In Fig. 11 we investigate the DNP, leakage current and energy levels in the regime, where the HF singlet-triplet rates dominate in magnitude over the triplet-triplet and inelastic rates close to the level crossing (13) and see Fig. 13.

Current hysteresis is a natural consequence of two stable DNP solutions for $T < T_{c,ST}$ — just as for the crossing of the triplets. For instance, if one increases the magnetic field from, say, $g_{\mu}B = 120 \mu eV$ for $T < T_{c,ST}$ [Fig. 11(a)], then the DNP will remain on the lower DNP solution until the critical field of about $g_{\mu}B \approx 140.9 \mu eV$, where the lower branch cease to exist. At this point, the DNP jumps discontinuously to the upper stable branch, such that the current also changes discontinuously as seen in Fig. 11(b). Likewise, when sweeping the field backwards from a high value of $g_{\mu}B$, then a discontinuum jumps is found in the current at the point, where the upper stable DNP cease to exist.

The stability of the DNP solution does not follow di-
FIG. 11. (Color online) The nuclear polarization $P$, leakage current $I/I_L$, and energy levels versus positive external magnetic field $g\mu_B B$ (in energy units) close to the singlet-triplet crossing for temperatures $T = 0.5T_{c,ST} < T_{c,ST}$ (a–c), $T = T_{c,ST}$ (d–f), and $T > T_{c,ST}$ (g–i). For $T < T_{c,ST}$, we observe two stable DNP values (black full lines) and an unstable one (blue dashed line) in Fig. (a), which leads to the hysteretic leakage current as seen in Fig. (b). The corresponding energy levels are seen in Fig. (c), where only the case of sweeping the magnetic field forward is shown for clarity. For $T \geq T_{c,ST}$, the DNP is single valued [Fig. (d,g)] such that no hysteretic current appears, see Fig. (e,h). Note the difference in the vertical scales between the DNP in Fig. (a) and Fig. (d,g). The vertical dashed black line indicates the simultaneous crossing of (i) the triplet energy $E_{T_+}$ (red full line) with the singlet energy $E_{S_+}$ (blue dashed line) and (ii) $E_{T_-}$ (green dashed line) with $E_{S_-}$ (brown full line). The current is seen to peak at the level crossing – essentially due to the enhanced HF singlet-triplet rate, which lifts the SB. The inset of Fig. (f) shows the non-monotonous energy level variation close to the crossing of $E_{T_+}$ and $E_{S_+}$. In general, $T_{c,ST}$ depends on several parameters of the system (see the main text). For the numerical example seen here, we find $k_B T_{c,ST} \approx 2.80 A_+\mu$, which is about one order of magnitude larger than $k_B T_{c,ST} \approx A_+\mu$. The parameters used here are: $A_+ = 80\mu V$, $A_R = 70\mu V$, $t = 100\mu V$, $\gamma_{th} = 1\mu V$, $T = 100K$ (i.e. $\gamma_{RL} = 1$), $h\Gamma_L N = 10^8\mu V$ and the dimensionless inelastic escape rate is chosen to be $\gamma_{in} = 10^{16}/T_L = 10^{-7}$, such that the HF rates dominates close to the singlet-triplet crossing (see Fig. [13]).

rectly from the solution of the implicit equation $[14]$. To determine the stability of the DNP against small fluctuations, we numerically propagate the rate equations $[40,11]$ in time until a stationary solution is reached $[12]$. The solution of the simplified rate equations $[10,11]$ and the numerical solution of the full rate equations $[11,12]$ with all rates match extremely well. The results in Fig. 11(d–i) calculated in the two ways fit perfectly.

Next, we consider the regime of $T \geq T_{c,ST}$, Fig. 11(d–i). By increasing the $B$-field away from the crossing of the triplets at $B = 0$, the triplet-triplet rates decrease, while the singlet-triplet rates increase, since the triplet energy $E_{T_+}$ ($E_{T_-}$) approaches the singlet energy $E_{S_+}$ ($E_{S_-}$) from below (above) [Fig. 11(d–i)]. Therefore, two new processes come into play to lift the SB, namely $T_+ \rightarrow S_+$ and $T_- \rightarrow S_-$, see Fig. 10. The closer the singlet and triplet levels are, the more effective are these two new processes, which in turn produce a leakage current peak at the singlet-triplet level crossing as seen in Fig. 11(e,h). Moreover, the pure triplet occupations $n_{T_+}$ decrease close to the singlet-triplet level crossing as a consequence of the enhanced triplet-singlet processes as seen in Fig. 14(a). Simultaneously, the occupation of the mixed triplet $T_\pm$ peaks at the level crossing. The reason is that the DNP decreases such that the escape rate $\Gamma_{D,L} \sim p^2 E_{T_\pm}$ becomes heavily suppressed as seen in Fig. 13(c).

The two dominant HF processes close to the singlet-triplet crossing, $T_+ \rightarrow S_+$ and $T_- \rightarrow S_-$, polarize the nuclei in opposite directions. When approaching the singlet-triplet crossing from below ($E_{T_+} < E_{S_+}$),
the DNP decreases [Fig. 11(d,g)]. This is consistent with the fact that the negatively-polarizing phonon-emission process \( T_- \rightarrow S_- \) is larger than the positively-polarizing phonon-absorption process \( T_+ \rightarrow S_+ \) as seen in Fig. 13(a). The DNP is seen to increase again once, the magnetic field \( g\mu_B B \) is tuned beyond the singlet-triplet crossing (indicated by the dashed vertical line in Fig. 11).

Interestingly, a very sharp – yet continuous – increase in the DNP is seen for \( T = T_{c,ST} \) at a higher magnetic field than the one at which the singlet and triplet levels cross, see Fig. 11(d). This behavior is qualitatively different from the one observed for the crossing of the triplets. In that case, the sharp increase in DNP is found at the same magnetic field as the one where the triplets cross; compare Fig. 7(d) to Fig. 11(d). In both cases, the sharp DNP increase is a precursor of the DNP bistability. The sharp DNP increase at a \( g\mu_B B \) beyond the singlet-triplet level crossing, is also reflected in the sudden increase of level splitting just after the level crossing as seen in the inset of Fig. 11(f). The mismatch between the level crossing and the sharp DNP increase indicates that triplet-singlet processes are not the only important ingredient close to the singlet-triplet crossing – although their rates dominate in magnitude. The triplet-triplet transitions also play a role. In fact, it is the inclusion of the triplet-triplet rates in the simplified rates (40-41) that leads to an enhancement of the transition temperature.

V. MONTE CARLO SIMULATIONS AND THE BREAKDOWN OF THE RATE EQUATION APPROACH

Now the rate equation approach is shown to be consistent with Monte Carlo simulation including an inelastic escape mechanism. We pay special attention to the case without the inelastic escape mechanism, where the rate equation approach is shown to break down. In this case, the Monte Carlo simulations show that no polarization can be induced by the leakage current as expected.

A. Breakdown of the rate equation description without the inelastic escape mechanism

If HFI is the only mechanism lifting SB, then the average DNP does not change. Now, we show that this
This shows that if $\Gamma^{\text{inc}} = 0$, then the quantity $n_{T_+} - n_{T_-} + (N/2)P + (1/2)[n_{\uparrow} - n_{\downarrow}]$ is conserved in the time evolution of the rate equations. In other words, for $\Gamma^{\text{inc}} = 0$, the stationary state of the rate equations depends on the initial occupations $n_{T_+}$ and $n_{T_-}$, which is unphysical. Thus, the rate equation description \cite{10.1038/nphys3257} breaks down for $\Gamma^{\text{inc}} = 0$. This is the basic problem with the dynamics presented in Ref. \cite{22}. However, for $\Gamma^{\text{inc}} \neq 0$ as used in this paper, the quantity $n_{T_+} - n_{T_-} + (N/2)P + (1/2)[n_{\uparrow} - n_{\downarrow}]$ is not conserved, which is evident from Eq. (46).

\section*{B. Monte Carlo simulations}

For $\Gamma^{\text{inc}} \neq 0$ – even if it is very small – the rate equations \cite{10.1038/nphys3257} gives a reliable description of the DNP in the SB regime. To validate this, we have performed Monte Carlo simulations leading to the same results.

The idea of the Monte Carlo simulation is – in some sense – to carry out a numerical experiment. The simulation is begun by placing the system in some initial state, say $|T_p\rangle$, with some initial polarization $P(t = 0)$. Thereafter, the system is updated in discrete time steps. From each state $|i\rangle$, there is a certain probability $p_{f,i}$ to go to another state $|f\rangle$ of the system within a single time step. We use a computer-generated random number to decide, if the system goes to another state or simply remains in the same state in a time step. The probability for a certain transition in a time step is proportional to its rate. A HF transition changes the nuclear polarization for the next time step, and, in turn, also the transition probabilities. Thus, the polarization dynamically changes in time along with the probabilities during the simulation. At some point in time, the polarization is such that the system has found a stationary state on the average. In order to get average properties, that can be compared to the results of the rate equation approach, we need to time average over the fluctuations of the simulation.

Appendix C gives more details on implementing the Monte Carlo simulations and shows examples of the DNP in single Monte Carlo simulations with and without the inelastic escape mechanism, respectively, in Figs. 15-17.

The main difference between the rate equation approach and the Monte Carlo approach is that the rate equations solely deal with average quantities. Therefore, the rate equations allow in some sense many processes to take place on average side by side. In contrast, the system is in a specific state in each instant of time during a Monte Carlo simulation. Both approaches neglect all quantum mechanical coherences in the description.

In Fig. 16, we see that the rate equation description and the Monte Carlo simulations agree for the DNP versus $T$ close to the crossing of the triplets. In the same way, we find excellent agreement between the two methods for finite $B$-fields close to the crossing of the triplets, and for $B$-fields around the single triplet crossings.

Furthermore, we have performed Monte Carlo simulations in the case of zero inelastic escape probability, $p^{\text{inc}} = 0$, where the rate equation approach for the DNP breaks down. We find that if $p^{\text{inc}} = 0$, then the time-averaged polarization is simply equal to the initial polarization of the simulation. See Fig. 16 for an example and its caption for a discussion. These simulations therefore confirm that no finite DNP is built up on average for HFIs being the only mechanism lifting SB, as expected \cite{10.1038/nphys3257}.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{occupations.png}
\caption{(Color online) (a) Occupations of the triplets $n_{T_+} = n_{T_+}$ (red full line) and $n_{T_-}$ (purple dashed line). (b) Occupations of the singlets, $n_{S_+}$ (orange dotted line) and $n_{S_-}$ (brown full line), are seen to differ slightly as anticipated in Eq. (12). (c) Occupations of the one-electron states $n^1_{\uparrow} (black full line)$. The vertical dashed black line indicate the position of the singlet-triplet level crossing. We observe a decrease in the occupations of the pure triplets $T_2$ close to the level crossing due to the enhanced HF singlet-triplet rates in this region, see Fig. (13a). In contrast, the occupation of the triplet $T_0$ increases heavily due to the DNP. The rate equations (A1-10) to obtain}
\end{figure}
In summary, we have analyzed the DNP and leakage current through a DQD in the SB regime due to a competition between HFIs and another inelastic escape mechanism from the triplets. We have demonstrated in detail how the DNP becomes bistable for temperatures $T$ below the transition temperature around both (i) the crossing of the three triplet levels and (ii) the crossing of the triplets $T_{\pm}$ with the singlets $S_{\pm}$. The bistable DNP leads naturally to hysteresis in the leakage current. We have found that the transition temperature for the crossing of the triplet levels, $T_{c,TT}$, is generally different from the transition temperature for the singlet-triplet crossing, $T_{c,ST}$. Moreover, $T_{c,TT} < T_{c,ST}$ for experimentally relevant parameters and the difference can be sizable, e.g. an order of magnitude. This enhancement of $T_{c,ST}$ stems from an interplay between the triplet-triplet and singlet-triplet HF rates, even though the latter often dominates by at least an order of magnitude. For $T_{c,TT} < T < T_{c,ST}$, current hysteresis appears around the singlet-triplet crossings at finite magnetic field, but is absent close to $B = 0$. Moreover, we found analytically $T_{c,TT} = (A_L + A_R)/(8k_B)$, where $A_L(R)$ is the effective HF constant of the left (right) dot. In contrast, $T_{c,ST}$ depends on various parameters, e.g. the inhomogeneity of the Overhauser field. Realistic HF constants of about $100\,\text{µeV}$ gives $T_{c,TT} \sim 300\,\text{mK}$, which is within experimental reach. Due to the enhancement of $T_{c,ST}$ compared to $T_{c,TT}$, it might be harder to observe due to the broadening of the Coulomb blockade peaks. However, this depends heavily on the actual parameters and experimental setup (see Tables C for details).

VI. SUMMARY, DISCUSSION AND OUTLOOK

In summary, we have modeled the DNP of the nuclear spins as a single valued quantity, $P$, as in e.g. Refs. \cite{33,34,35,36,37} and \cite{38}. In reality, the polarization will vary in space leading to a more complex behavior, which is more involved to model. An extension of the model could be to use different DNPs for each dot. In such an approach, it is an open question, if the two DNPs would become bistable at the same transition temperature or not. Moreover, as emphasized in the paper, the difference in the Overhauser field between the dots is important to
FIG. 17. (Color online) (a) An example of a Monte Carlo simulation showing that the nuclear polarization (blue full line) is not changed on the average due to the leakage current when only HFIs lift the SB, i.e. $\hat{H}^{\text{inel}} = 0$. The magnetic field is tuned close to the singlet-triplet crossing leading to a level ordering as seen in (b). Moreover, we choose a very low temperature such that finite DNP could appear for $\hat{H}^{\text{inel}} \neq 0$. The initial polarization, $P(t = 0) = 0$ (black dashed line), is found to be equal to the time-averaged polarization, $\bar{P} \simeq 0.002$, within the uncertainty. Similarly, the average occupations in the simulation are found to be $\bar{\pi}_{T_+} \simeq 0.26$, $\bar{\pi}_{T_-} \simeq 0.15$, $\bar{\pi}_{S_+} \simeq 0.48$, $\bar{\pi}_{S_-} \simeq 0.0663$, $\bar{\pi}_{\sigma,S_+} \simeq 0.063$ and $\bar{\pi}_{\sigma} \simeq \pi_{\sigma} \simeq 0.017$. Thus, in contrast to the $\hat{H}^{\text{inel}} = 0$ case, we find $\bar{\pi}_{T_+} \neq \bar{\pi}_{T_-}$. The parameters are: $A_R = 50\mu eV$, $A_L = 30\mu eV$, $t = 50\mu eV$ and $\gamma_{ph} = 5\mu eV$ such that the singlet and triplet energy levels are well separated, i.e. $t \gg \gamma_{ph}$. Moreover, we use $p_L = p_T = 0.45$, $p_D = 0.0065$ and $g_{HF} = 2$. (b) An illustration of why no nuclear polarization is expected to be induced, when HFI is the only mechanism lifting SB. In contrast, escape from the triplets, which compete with the HF rates, $\sigma^{\text{inel}}$, is a valid approach as long as the inelastic rate is smaller than the dominant HF rate close to the level crossing, as studied here. Nevertheless, the inelastic rates can be increased experimentally, e.g. by choosing a material with strong spin-orbit coupling or by tuning the levels compared to the chemical potentials of the leads, so co-tunneling becomes more probable. In such cases, it could be interesting to repeat our analysis including the energy dependences of the co-tunneling rates and/or spin-orbit rates. Future work could also analyze the effects of including a more detailed description of the phonons. However, we believe that the essential physics is captured by including the asymmetry between energy emission and absorption in the HF rates.

We have studied in detail the case of a single constant inelastic escape rate from the triplets, which compete with the HF rates. Neglecting the energy dependence of the inelastic rates is a valid approach as long as the inelastic rate is smaller than the dominant HF rate close to the level crossing, as studied here. Nevertheless, the inelastic rates can be increased experimentally, e.g. by choosing a material with strong spin-orbit coupling or by tuning the levels compared to the chemical potentials of the leads, so co-tunneling becomes more probable. In such cases, it could be interesting to repeat our analysis including the energy dependences of the co-tunneling rates and/or spin-orbit rates. Future work could also analyze the effects of including a more detailed description of the phonons. However, we believe that the essential physics is captured by including the asymmetry between energy emission and absorption in the HF rates.

VII. ACKNOWLEDGMENTS

We are especially thankful to János Asbóth and András Pályi for insightful discussions on the nature of non-zero nuclear polarization in the SB setup. Moreover, we thank Andrea Donarini, Sigmund Kohler, Rafael Sánchez, Gerold Kiesslich, Marla Prada, Jeroen Danon, Jesús Iñarrea and Fernando Domínguez for useful discussions. AML acknowledges the Juan de la Cierva program (MICINN) and the Carlsberg Foundation. AML, CLM and GP acknowledge Grant No. MAT2011-24331 and the ITN Grant 234970(EU). LLB and IV acknowledge Grant No. FIS2011-28838-C02-01. We acknowledge FIS2010-22438-E (Spanish National Network Physics of Out-of-Equilibrium Systems).

Appendix A: Expressions for the full rate equations of the model

In the main paper, we give the full set of rate equations including all non-zero rates in a compact form in Eq.(9). For completeness, we provide here the detailed equations
as illustrated in Fig. 4

Appendix B: Current expressions close to the crossing of the triplet levels for non-equal inelastic escape rates

Throughout this paper, we have considered the case of equal and constant inelastic escape rates \( \Gamma_{\text{ine}} \) from the triplet states \( T = T_+, T_p \), see Eq. (12) \[\text{[22]}\] However, depending on the inelastic escape mechanism, the rates might be different. For completeness, we discuss this briefly in this appendix in a particularly simple case.

Here we consider the case, where the inelastic escape rates are invariant under the same exchange of indices as the tunneling rates Eq. (15), i.e.

\[ \Gamma_{\text{ine}, T_p} = \Gamma_{\text{ine}, T_+}, \quad \Gamma_{\downarrow, T_+} = \Gamma_{\downarrow, T_-}, \quad \Gamma_{\uparrow, T_+} = \Gamma_{\uparrow, T_-}. \]  

(B1)

Following the same steps leading to the polarization equation in Sec. III.A, we find that \( n_\downarrow = n_\downarrow \) and \( n_{T_+} = n_{T_-} \) still hold true such that the implicit equation (24) for the polarization,

\[ W_{T_+, T_+} = W_{T_+, T_-} = W_{T_-, T_+} = W_{T_-, T_-}, \]  

(B2)

and the transition temperature, \( k_B T_{\text{tr}} = A_+ / 4 \) \[\text{[27]}\], remain unchanged compared to the main text. Furthermore, \( n_{S_+} = n_{S_-} \) and Eqs. (29) and (30) also still hold true, whereas the explicit expressions for the occupations (31) and the current expressions for the occupations (32) are changed slightly. In the numerator of \( n_{T_\pm} \) (31b), the rate \( \Gamma_{\text{ine}, T_\pm} \) is replaced by \( \Gamma_{\text{ine}, T_\pm} \). Similarly, in the numerator of \( n_{T_\pm} \) (31a), one has to make the replacement \( 2\Gamma_{\text{ine}} \rightarrow \Gamma_{\text{ine}, T_+} + \Gamma_{\text{ine}, T_-} \). The common denominator of the occupations (31) changes to \( \Lambda = 2\Gamma_{T_\pm, T_\pm} + \Gamma_{T_+, T_+} + \Gamma_{T_+, T_-} + 3\Gamma_{T_-, T_+} + \Gamma_{T_-, T_-} \).

Thus, the current for \( p = 0 \) still depends on the HF rate \( W_{T_+, T_+} \) in general.

In summary, when the inelastic rates have the same invariances under interchange of indices as the tunneling rates, then the polarization equation (22) does not change and the current expression changes only slightly.

\[ I(p = 0) = \frac{8\Gamma_{\text{ine}, T_+}}{\sum} \left[ W_{T_+, T_+} \Gamma_{\uparrow, T_+} + W_{T_p, T_p} + 8\Gamma_{\text{ine}, T_+} \right] \]  

(B4a)

where \( T_\pm \) was used for simplicity and

\[ \sum = W_{T_+, T_+} \left[ 6\Gamma_{\text{ine}, T_+} + 6\Gamma_{\text{ine}, T_-} + 9\Gamma_{L_\pm} \right] \]  

(B4b)
However, if the inelastic rates are invariant under other interchange of indices, then the polarization condition \(2\vec{P} > N\) and the transition temperature might change.

**Appendix C: Details on the implementation of the Monte Carlo simulations**

We implement all the possible transitions between the states \(\{↑, ↓, T_s, T_p, T_{-}, S_+, S_-\}\) as shown graphically on Fig. \(3\). Therefore, we are not limited to simulate a specific level crossing. For the transition probabilities in a single time step, we use the same functional dependences as for the rate expressions \(\sum_{j}^{16,17}\), since rates and probabilities are proportional. In the formulas \(\sum_{j}^{16,17}\), we exchange the rates \(\Gamma_{R(L)}\) by \(p_{R(L)}\) and \(\Gamma^{\text{inc}}\) by \(p^{\text{inc}}\).

For instance, \(p_{T_p,↑} = p_{L}/(2N^2)\) is the probability for going to \(T_p\) given that the system is in the one-electron state \(\uparrow\). Likewise, we exchange the factor \(1/(2N)\) in the HF probabilities \(\sum_{j}^{16,17}\) by the parameter \(\eta_{HF}\) in the HF probabilities. We therefore can tune the magnitude of the HF transition probabilities compared to the inelastic transition probabilities. Thus, we can easily study the same physical situation as in the rate equation approach. For instance, Figs. \(5\) and \(16\) both study large singlet-triplet energy separation and zero magnetic field.

To minimize the computational load, we choose the transition probabilities within a single time step as high as possible, such that the system does not remain in the same state over too many time steps. This can be understood as a long physical time duration for each time step. Nevertheless, we have to choose numbers such that the sum of all probabilities for leaving a specific state is always smaller than one in each time step, e.g. for \(T_p\) this amounts to

\[
2p^{\text{inc}} + p_{\uparrow,T_p} + p_{T_p} + p_{R(L)T_p}(t) + p_{R(L)T_p}(t) < 1. \quad (C1)
\]

In this way, the possibility of staying in the same state (here \(T_p\)) within a time step remains in the simulation. In the real experiment, the polarization change by \(dP = \pm 2/N\) due to a single HF transition. In the simulation, however, \(dP\) is increased substantially in order to obtain faster convergence to a stationary polarization. We emphasize that the choice of \(dP\) does not affect the value of the stationary polarization, but it does indeed affect the typical fluctuations around this value. Thus, an artifact of choosing \(dP\) larger than \(2/N\) is the artificially increased fluctuations around the stationary polarization – as seen in Figs. \(15\) and \(17\) – compared to the experimental situation. However, since we are only interested in average values, this is not a concern here. Choosing \(dP\) therefore is a compromise between maximizing convergence time and minimizing fluctuations.

In order to find the stationary DNP, we choose a \(dP\), perform the Monte Carlo simulation a number of times for a given initial DNP and then average over the results. The averaging makes it easier to decide in a computationally cheap way, if convergence is reached. To make sure that the found stationary DNP \(\bar{P}\) is stable, we show that an initial DNP \(P(t = 0) > \bar{P}\) decreases versus time and that an initial DNP with \(P(t = 0) < \bar{P}\) increases versus time. We stress that the stationary DNP can also be found from doing the single Monte Carlo simulation as seen in Fig. \(15\) but it requires a much smaller \(dP\). Moreover, the fluctuations in DNP increase with temperature, since higher \(T\) increases the phonon-absorption HF transition probabilities, which increases the number of likely transitions.

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1. R. Hanson, L. P. Kouwenhoven, J. R. Petta, S. Tarucha, and L. M. K. Vandersypen, Rev. Mod. Phys. 79, 1217 (2007).
2. W. G. van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha, and L. P. Kouwenhoven, Rev. Mod. Phys. 75, 1 (2002).
3. S. M. Reimann and M. Manninen, Rev. Mod. Phys. 74, 1283 (2002).
4. L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Reports on Progress in Physics 64, 701 (2001).
5. K. Ono, D. G. Austing, Y. Tokura, and S. Tarucha, Science 297, 1313 (2002).
6. K. Ono and S. Tarucha, Phys. Rev. Lett. 92, 256803 (2004).
7. D. Weinmann, W. Häusler, and B. Kramer, Phys. Rev. Lett. 71, 984 (1995).
8. D. Weinmann, W. Häusler, and B. Kramer, Annalen der Physik 508, 652 (1996), ISSN 1521-3889.
9. M. Ciorga, A. S. Sachrajda, P. Hawrylak, C. Gould, P. Zawadzki, S. Jullian, Y. Feng, and Z. Wasilewski, Phys. Rev. B 61, R16315 (2000).
10. F. H. L. Koppens, J. A. Folk, J. M. Elzerman, R. Hanson, L. H. W. van Beveren, I. T. Vink, H. P. Tranitz, W. Wegscheider, L. P. Kouwenhoven, and L. M. K. Vandersypen, Science 309, 1346 (2005).
11. A. C. Johnson, J. R. Petta, J. M. Taylor, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Nature 435, 925 (2005).
12. A. C. Johnson, J. R. Petta, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Phys. Rev. B 72, 165308 (2005).
13. A. B. Vorontsov and M. G. Vavilov, Phys. Rev. Lett. 101, 226805 (2008).
14. K. Saito, S. Okubo, and M. Eto, Physica E 40, 1149 (2008), ISSN 1386-9477, 17th International Conference on Electronic Properties of Two-Dimensional Systems.
15. F. Qassemi, W. A. Coish, and F. K. Wilhelm, Phys. Rev. Lett. 102, 176806 (2009).
16. W. A. Coish and F. Qassemi, Phys. Rev. B 84, 245407 (2011).
17. A. V. Khatskii and Y. V. Nazarov, Phys. Rev. B 61, 12639 (2000).
18. V. N. Golovach, A. Khatskii, and D. Loss, Phys. Rev.
Here the Zeeman energy splitting between the nuclear spins states is negligible compared to the one of the electronic states. This is because the nuclear magneton is about 2000 times smaller than the Bohr magneton due to the mass ratio of the electron and the proton.

For very low temperatures compared to the triplet energy, the singlet-triplet emission rates become comparable. Here we find DNP on the order of 60%.

We remark that hysteretic behavior due to DNP is not limited to quantum dot systems, but have also been found experimentally e.g. in quantum Hall systems.

Formally, this is equivalent to including the inter-dot tunnelling and the Overhauser field in usual degenerate perturbation theory for the chosen states.

The depolarization can be described by including a phenomenological term $-\frac{W_{p}}{\gamma_{N}}P$ in the DNP rate Eq. (10).

In order for the triplet-triplet absorption and singlet-triplet emission rates to be comparable, here we have to much lower than the triplet level splitting $E_{T} - E_{S}$.

To be sure, thus, they can be comparable to the emission singlet-triplet rates, which in turn can cause the approximation used here to fail. However, due to the difference between the overall prefactors $(W_{T_{S}} \approx A_{T}^{2}$ and $W_{T_{S}} \approx A_{T}^{2}$), $k_{T}$ indeed has to be very much lower than the triple-tetat level splitting $|E_{T_{p}} - E_{p}|$ in order for the triplet-triplet absorption and singlet-triplet emission rates to be comparable. Here we do not consider such very low temperatures.

Note for instance that if $E_{T_{p}} > E_{T_{S}}$ and we take

$W_{T_{p},T_{s}} \approx \propto n_{D}(E_{T_{p}} - E_{T_{p}}) + 1$ and $W_{T_{S},T_{s}} \approx \propto n_{D}(E_{T_{p}} - E_{T_{p}}) + 1$.

The depolarization can be described by including a phenomenological term $-\frac{W_{p}}{\gamma_{N}}P$ in the DNP rate Eq. (10).

For very low temperatures compared to the triplet energy splitting, the phonon absorption triplet-triplet rates become heavily suppressed (see Sec. II C 3). Thus, they can become comparable to the emission singlet-triplet rates, which in turn can cause the approximation used here to fail. However, due to the difference between the overall prefactors $(W_{T_{S}} \approx A_{T}^{2}$ and $W_{T_{S}} \approx A_{T}^{2})$, $k_{T}$ indeed has to be very much lower than the triple-tetat level splitting $|E_{T_{p}} - E_{p}|$ in order for the triplet-triplet absorption and singlet-triplet emission rates to be comparable. Here we do not consider such very low temperatures.

Note for instance that if $E_{T_{p}} > E_{T_{S}}$ and we take $W_{T_{p},T_{s}} \approx \propto n_{D}(E_{T_{p}} - E_{T_{p}}) + 1$ and $W_{T_{S},T_{s}} \approx \propto n_{D}(E_{T_{p}} - E_{T_{p}}) + 1$.
Thus, in this respect, the DNP becomes model independent. Here \( n_B(E) = \frac{e^{E/k_B T} - 1}{e^{E/k_B T} + 1} \) is the Bose function.

125 S. Strogatz, *Nonlinear dynamics and chaos: with applications to physics, biology, chemistry, and engineering* (Perseus Books, 1994), 1st ed.

126 This can, for instance, be done by using that Eq. (26) for \( B = 0 \) goes from having one to three solutions for \( P \), when the derivative of \( \tanh[A_P P/(4k_B T)] \) at \( P = 0 \) is one.

127 Remember that all the non-zero HF rates between the triplets become equal in steady state due to the implicit polarization equation (24), and therefore the final and initial state is not emphasized in the notation of \( w \).

128 Note that there is no need to choose the rate \( \Gamma_L \) explicitly in these plots. However, to specify the dimensionless hyperfine rate \( w = W_{T \rightarrow T}/\Gamma_\perp \), one needs to give a number for \( \hbar \Gamma_L N \), where \( N \) is the total number of nuclear spins in the DQD, around \( 10^4 - 10^7 \) depending on the material etc. Choosing \( \hbar \Gamma_L N = 10^4 \mu eV \) corresponds to – for instance – having \( \hbar \Gamma_L \sim 100 \mu eV \) and \( N \sim 10^5 \).

129 The lowest order term in the expansion in \( \gamma_{\text{ine}} \) gives the same for \( \gamma_{\perp \perp} \neq 1 \).

130 N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Thomson Learning, 1976).

131 For \( g \mu_B B < 0 \), the energy levels that cross are reversed, i.e. \( E_{T_+} \) cross \( E_{S_+} \) and \( E_{T_-} \) cross \( E_{S_-} \). This means that one has to neglect other HF rates in order to construct a simple model describing the level crossing for \( g \mu_B B < 0 \). However, this is straightforward following the same principles as outlined in the main text.

132 We note that this approach fails for the parameters considered here, e.g. \( A_L = 80 \mu eV \), \( A_R = 70 \mu eV \), \( t = 100 \mu eV \), \( \gamma_{\text{ph}} = 1 \mu eV \), \( \Gamma_L = \Gamma_R \), \( \hbar \Gamma_L N = 10^6 \mu eV \) and \( \gamma_{\text{ine}} = 10^{-7} \). Nevertheless, the approximation becomes better (but far from perfect) very close to the singlet-triplet crossing by increasing \( t \) to \( t = 10^4 \mu eV \), since this decreases the influence of the triplet-triplet HF rates. However, we are not interested in such extreme cases of very large \( t \).

133 To see that \( G(P) > 0 \) and \( G(P) \neq 0 \), we begin by noting that \( G(P) \) is a sum of products of rates, which are all greater than or equal to zero. Thus, \( G(P) \geq 0 \). However, at least one of these terms is always strictly positive, namely \( \Gamma_{T+} \). Hence, \( G(P) > 0 \).

134 We choose slightly different parameters in Fig. 11 compared to Fig. 7. Using the parameters of Fig. 7, a very similar transition temperature is found, \( T_{c,ST} \approx 2.81 A_+/k_B \), – even though the inelastic rate dominates for magnetic fields close to the singlet-triplet crossing.

135 We remark that close to the critical temperature in Fig. 16 it is hard to obtain conclusive average polarization values of the Monte Carlo simulations due to fluctuations.