Nuclear spin pumping and electron spin susceptibilities

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In this work we present a straightforward formalism to evaluate the nuclear spin dynamics driven by hyperfine interaction with non-equilibrium electron spins. To describe the dynamics up to second order in the hyperfine coupling, it suffices to evaluate the susceptibility and fluctuations of the electron spin. Our approach does not rely on a separation of electronic energy scales or the specific choice of electronic basis states, thereby overcoming practical problems which may arise when using a more traditional formalism based on rate equations.

INTRODUCTION

In recent years, considerable theoretical and experimental work is aimed at the controlled manipulation of electron spins in nanoscale solid state devices [1]. This research is motivated by actual applications, such as in digital information storage and read-out [2, 3], but also by the possibility of using the spin of electrons as computational units (qubits) in a quantum computer [4].

One of the mechanisms influencing the electron spin dynamics in these nano-devices is the hyperfine interaction between the electron spin and the nuclear spins of the device's constituent material. For spin qubits hosted in semiconductor quantum dots [5], hyperfine interaction has been identified as the main source of decoherence, causing the spin coherence time to be in the ns range [6–8]. Much recent experimental and theoretical work is aimed at suppressing this hyperfine induced decoherence [9, 10]. In other semiconductor nanostructures [11, 12] — possibly also metallic structures [13] — hyperfine interaction can even dominate the electronic transport properties and spin dynamics. Understanding the role of hyperfine interaction in spintronic devices therefore is crucial in the development of the field.

Traditionally, the interplay between electronic and nuclear spins is treated in the convenient framework of rate equations [14, 15]. The rates of hyperfine transitions flipping nuclear spins 'up' and 'down' are separately calculated using Fermi's golden rule, and the balancing of these rate yields the net nuclear spin pumping in the system. Although this approach works very well in many cases [16, 17], it can become cumbersome when the electron spin dynamics are more complicated. Strong dynamical nuclear spin effects have been observed in GaAs double quantum dots in the spin blockade regime under conditions of electron spin resonance [18]. Nuclear spin flips in this setup are due to second order processes, and some of the transitions have a vanishingly small energy difference between initial and virtual state, which cannot be dealt with in a standard Fermi golden rule approach [19]. The situation is even worse for systems with strong spin-orbit coupling. In InAs nanowire double quantum dots in the spin blockade regime, signatures of strong dynamic polarization have been observed too [20]. In this case, the presence of too many comparable energy scales makes it impossible to choose a unique set of basis states to describe the electron dynamics in [21]. It is clear that a basis-independent description of the coupled electron-nuclear spin dynamics, not involving any separate transition rates, is highly desirable.

In this work we present an alternative way to evaluate the spin dynamics of electrons and nuclei coupled to each other by hyperfine interaction. We show that, in order to describe the nuclear spin dynamics up to second order in the hyperfine coupling, it suffices to evaluate the fluctuations and susceptibility of the electron spins in the system, which can be done using linear response theory. Our approach does not rely on the calculation of separate transition rates with Fermi's golden rule, nor on the choice of electronic basis states to work in. As a result, the formalism is applicable in cases where Fermi's golden rule cannot be used, and is thus better and more general.

Since a Fermi's golden rule (FGR) calculation often has to be adapted to specific circumstances [14–17], it is difficult to pick a single implementation of it and make a good comparison to our approach. Therefore we will illustrate our ideas with two example systems: (i) We will show how in a simplest toy system our formalism and a smart implementation of FGR produce identical results. (ii) We will investigate a more complicated system, which cannot be dealt with in an FGR approach, and show how our formalism straightforwardly produces an equation for the hyperfine driven nuclear spin dynamics.

MAIN RESULT

Let us start with presenting the main result of our work: an equation for the hyperfine driven dynamics of the expectation value of a nuclear spin coupled to (many) electronic spins. Under conditions which we will specify below, this equation takes the closed form

$$\frac{d\langle \hat{K}^a \rangle}{dt} = \varepsilon^{abc} S^b \langle \hat{K}^c \rangle - \frac{1}{2} Q^{ab} \langle \hat{K}^b \rangle + \frac{1}{3} P^a, \qquad (1)$$

where $\{a,b,c\} \in \{x,y,z\}$, and we use the convention that over repeated indices still has to be summed. The operator $\hat{\mathbf{K}}$ is the nuclear spin operator for the nucleus under consideration, and ε^{abc} is the permutation tensor. The vectors $\mathbf{S}$ and $\mathbf{P}$, and the matrix Q are defined as

$$\begin{aligned} S^a &= Av_0\langle \hat{S}^a(\mathbf{r}_n,t)\rangle \\ Q^{ab} &= (Av_0)^2(\delta^{ab}R^{cc} - R^{ba}) \\ P^a &= (Av_0)^2 K(K+1)\varepsilon^{abc}\chi^{bc}, \end{aligned} \tag{2}$$

A being the material-specific hyperfine coupling energy, $1/v_0$ the density of nuclei, K the total nuclear spin, $\mathbf{r}_n$ the position of the nucleus, and $\hat{\mathbf{S}}$ the electron spin density operator, $\hat{\mathbf{S}}(\mathbf{r},t) = \hat{\psi}^\dagger_\alpha(\mathbf{r},t)\boldsymbol{\sigma}_{\alpha\beta}\hat{\psi}_\beta(\mathbf{r},t)$. Note that we have set for transparency $\hbar = 1$, or, in other words, we express all energies in terms of corresponding frequencies. The symbols χ and R represent correlation functions of the local electronic spin density,

$$\begin{aligned} R^{ab} &= \int^t \langle [\hat{S}^a(\mathbf{r}_n,t), \hat{S}^b(\mathbf{r}_n,t')]_+\rangle_c dt' \\ \chi^{ab} &= -i\int^t \langle [\hat{S}^a(\mathbf{r}_n,t), \hat{S}^b(\mathbf{r}_n,t')]_-\rangle_c dt', \end{aligned} \tag{3}$$

where the square brackets denote the (anti)commutator of two operators, i.e., $[\hat{A},\hat{B}]_\pm = \hat{A}\hat{B} \pm \hat{B}\hat{A}$, and the subscript c means that we have to use the 'connected' part of the expectation value, i.e., remove the contribution of the averages $\langle AB\rangle_c = \langle AB\rangle - \langle A\rangle\langle B\rangle$. Note that, since hyperfine interaction works two-way, the electronic correlation functions, and thereby $\mathbf{S}$, Q and $\mathbf{P}$, are in turn also affected by the state of the nuclear spins in the system. This creates a feedback mechanism in Eq. (1), which makes the equation non-linear. In all practical implementations, one can make use of the large difference in time scales of the electronic and nuclear spin dynamics. This allows to treat the nuclear spins as a static 'classical' magnetic field when evaluating the electronic correlators.

The first term in Eq. (1) is first order in the hyperfine coupling A and gives rise to a precession of the nuclear spin around the direction of the time-averaged local electron spin polarization. The precession frequency depends on the magnitude of this polarization $|v_0\langle\hat{\mathbf{S}}(\mathbf{r}_n,t)\rangle|$, as well as on the strength A of the hyperfine coupling.

The other two terms are second order in A, and involve the exchange of angular momentum between electrons and the nucleus. The correlation functions R^{ab} in the matrix Q contain the 'classical' noise in the coordinates $\hat{\mathbf{S}}(\mathbf{r}_n,t)$ and, as dictated by the Onsager relations, lead to a relaxation of the nuclear spin coupled to these coordinates. The correlators χ^{ab} in the vector $\mathbf{P}$ correspond to the susceptibility in the same coordinates, and can give rise to nuclear spin pumping in the system. One can understand this by interpreting the χ^{ab} as the 'quantum' noise in the electron spin density: the part of the fluctuations in $\hat{\mathbf{S}}(\mathbf{r}_n,t)$ which solely exists due to the non-commutativity of the spin operators. By their quantum nature these fluctuations violate the Onsager relations, and can thus indeed drive the system out of equilibrium.

In order to find an explicit expression for $d\langle\hat{\mathbf{K}}\rangle/dt$ as given by Eq. (1), one needs to find the elements of $\mathbf{S}$, Q and $\mathbf{P}$. The first step is to find the steady-state electron spin density $\langle\hat{\mathbf{S}}(\mathbf{r}_n)\rangle$, which immediately yields the vector $\mathbf{S}$. The elements of Q and $\mathbf{P}$ correspond to the local fluctuations and susceptibility of this electron spin density, and can be evaluated using linear response theory, as we will illustrate below. Let us emphasize here that none of the steps in such a calculation involves the evaluation of any separate transition rate between different electronic-nuclear levels or depends on the specific choice of electronic basis states.

DERIVATION

We will now show how to derive Eq. (1) from a standard second order perturbation theory in the hyperfine interaction. We start from the second order time-evolution equation for the density matrix $\hat{\rho}$ of the whole coupled electron-nuclear spin system

$$\frac{d\hat{\rho}}{dt} = -i\left[\hat{H}_{\rm hf}(t),\hat{\rho}\right] - \int^t \left[\hat{H}_{\rm hf}(t),\left[\hat{H}_{\rm hf}(t'),\hat{\rho}\right]\right]dt', \tag{4}$$

$\hat{H}_{\rm hf}$ being the hyperfine Hamiltonian coupling all nuclear spins in the system to the local electron spin density. Assuming that the electrons of interest have s-type orbitals, we can write for the hyperfine Hamiltonian

$$\hat{H}_{\rm hf}(t) = Av_0\sum_n \hat{\mathbf{S}}(\mathbf{r}_n,t)\cdot\hat{\mathbf{K}}_n(t). \tag{5}$$

Making use of the large difference in time scales of the electronic and nuclear spin dynamics, we separate the two parts of the density matrix, $\hat{\rho} = \hat{\rho}_e \otimes \hat{\rho}_k$. We trace over the rapidly changing electronic degrees of freedom, assuming the nuclear part of the density matrix to be static on these time scales. This allows us to extract from Eq. (4) a separate time-evolution equation for $\hat{\rho}_k$, and thus derive equations of motion for the expectation values of the nuclear spin operators.

A single nucleus, $K = 1/2$

Let us first focus on the transparent case of one single nucleus with nuclear spin $K = 1/2$, interacting with a local electron spin density. Up to first order, i.e., using only the first term in (4), we find

$$\frac{d\langle\hat{K}^a\rangle^{(1)}}{dt} = {\rm Tr}\left\{\hat{K}^a\frac{d\hat{\rho}_k^{(1)}}{dt}\right\} = \varepsilon^{abc}S^b\langle\hat{K}^c\rangle, \tag{6}$$

describing a precession of the nuclear spin around the local electron spin density.

The exchange of angular momentum between electrons and the nucleus is to leading order described by the second term in (4), i.e., second order perturbation theory in $\hat{H}_{\rm hf}$. Applying the commutation relations for nuclear spin operators, and using that for spin-$\frac{1}{2}$ operators $[\hat{K}^a, \hat{K}^b]_+ = \frac{1}{2}\delta^{ab}$, a somewhat lengthy but straightforward calculation gives for the three elements $d\langle\hat{K}^a\rangle/dt$ the second order expression

$$\frac{d\langle\hat{K}^a\rangle^{(2)}}{dt} = \frac{(Av_0)^2}{2}\Big\{R^{ba}\langle\hat{K}^b\rangle - R^{bb}\langle\hat{K}^a\rangle + \varepsilon^{abc}\tfrac{1}{2}\chi^{bc}\Big\}, \quad (7)$$

which is, combined with Eq. (6), identical to the expression given in Eq. (1).

Many nuclei, $K > 1/2$

When following the same derivation for the case of many nuclei and spin higher than $K = 1/2$, it is generally not possible to derive a closed set of equations for $d\langle\hat{\mathbf{K}}_n\rangle/dt$. The first order equations do not change and are still given by Eq. (6). The second order correction however does change, and becomes

$$\begin{aligned}\frac{d\langle\hat{K}_n^a\rangle^{(2)}}{dt} = \frac{(Av_0)^2}{2}\Big\{&R^{ba}\langle\hat{K}_n^b\rangle - R^{bb}\langle\hat{K}_n^a\rangle \\ &+ \varepsilon^{abc}\chi_{nn}^{bd}\langle[\hat{K}_n^d, \hat{K}_n^c]_+\rangle \\ &+ 2\sum_{m\neq n}\varepsilon^{abc}\chi_{nm}^{bd}\langle\hat{K}_m^d\hat{K}_n^c\rangle\Big\},\end{aligned} \quad (8)$$

where the correlator χ has acquired two more indices,

$$\chi_{nm}^{ab} = -i\int^t \langle[\hat{S}^a(\mathbf{r}_n, t), \hat{S}^b(\mathbf{r}_m, t')]_-\rangle_c dt'. \quad (9)$$

This correlation function now describes the *non-local* susceptibility of the electron spin density, i.e., the linear response in the coordinate $\hat{S}^a$ at the position $\mathbf{r}_n$ due to a perturbation along $\hat{S}^b$ at another position $\mathbf{r}_m$.

We notice two problems which prevent us from deriving a closed set of equations from Eq. (8): First of all, for $K > 1/2$ we cannot make the convenient simplification $[\hat{K}_n^d, \hat{K}_n^c]_+ \propto \delta^{cd}$, and secondly also correlations $\langle\hat{K}_m^d\hat{K}_n^c\rangle$ between *different* nuclei apparently play a role.

Let us first address the issue concerning the averages of the form $\langle\hat{K}_m^d\hat{K}_n^c\rangle$. We start by splitting these correlators in a connected and an unconnected part, $\langle\hat{K}_m^d\hat{K}_n^c\rangle = \langle\hat{K}_m^d\hat{K}_n^c\rangle_c + \langle\hat{K}_m^d\rangle\langle\hat{K}_n^c\rangle$. The unconnected part of these correlators results in a contribution $(Av_0)^2\sum_m \varepsilon^{abc}\chi_{nm}^{bd}\langle\hat{K}_m^d\rangle\langle\hat{K}_n^c\rangle$, which actually is a correction to the first-order equation (6). The effective field around which $\langle\hat{\mathbf{K}}_n\rangle$ precesses is changed $S^a \to S^a + (Av_0)^2\sum_m \chi_{nm}^{ab}\langle\hat{K}_m^b\rangle$, i.e., it gains a term due to the electron spin mediated nuclear spin-spin coupling. We will focus on leading order effects only and disregard this correction, which means that we can interpret all brackets $\langle\ldots\rangle$ in (8) as indicating connected correlators [22].

We now investigate the dynamics of those correlators in more detail. We can derive for $\langle\hat{K}_l^a\hat{K}_m^b\rangle$ similar equations of motion as for the average $\langle\hat{K}_n^a\rangle$. Up to second order in Av_0 we find the two equations

$$\begin{aligned}\frac{d\langle\hat{K}_l^a\hat{K}_m^b\rangle^{(1)}}{dt} &= Av_0\Big\{\langle\hat{S}^c(\mathbf{r}_l, t)\rangle\varepsilon^{dac}\langle\hat{K}_l^d\hat{K}_m^b\rangle + \langle\hat{S}^c(\mathbf{r}_m, t)\rangle\varepsilon^{dbc}\langle\hat{K}_l^a\hat{K}_m^d\rangle\Big\} \\ \frac{d\langle\hat{K}_l^a\hat{K}_m^b\rangle^{(2)}}{dt} &= \frac{(Av_0)^2}{2}\Big\{(R_{ml}^{cd} + R_{lm}^{dc})\varepsilon^{adf}\varepsilon^{bce}\langle\hat{K}_l^f\hat{K}_m^e\rangle + R_{ll}^{cd}\varepsilon^{ace}\varepsilon^{edf}\langle\hat{K}_l^f\hat{K}_m^b\rangle + R_{mm}^{cd}\varepsilon^{bce}\varepsilon^{edf}\langle\hat{K}_l^a\hat{K}_m^f\rangle \\ &\quad + \sum_n\Big(\chi_{ln}^{cd}\varepsilon^{ace}\langle\hat{K}_n^d\hat{K}_l^e\hat{K}_m^b + \hat{K}_l^e\hat{K}_m^b\hat{K}_n^d\rangle + \chi_{mn}^{cd}\varepsilon^{bce}\langle\hat{K}_n^d\hat{K}_l^a\hat{K}_m^e + \hat{K}_l^a\hat{K}_m^e\hat{K}_n^d\rangle\Big)\Big\},\end{aligned} \quad (10)$$

where again all brackets indicate connected correlators. We see that hyperfine interaction does not *create* correlations between the different nuclear spins. The correlations will only start evolving when they already exist. We can thus make the practical assumption that the nuclear spins are initially in an uncorrelated thermal equilibrium state, which allows us to neglect the correlators of different nuclei, i.e., the last term in Eq. (8).

This assumption also solves the issue that the term $\langle[\hat{K}_n^d, \hat{K}_n^c]_+\rangle$ formally cannot be simplified further. Due to the small nuclear Zeeman energy, the thermal equilibrium state of the nuclear spin system is in all realistic cases in the high-temperature limit. If we further restrict ourselves to the build-up of small polarizations, we can approximate the nuclear spin density matrix to be nearly isotropic, resulting in $\langle\hat{K}_n^c\hat{K}_n^d\rangle \approx \frac{1}{3}\delta^{cd}K(K+1)$, where K is the total nuclear spin. We see that in that case Eq. (8) reduces to our main result, Eq. (1).

IMPLEMENTATION

With the formalism presented in Eqs (1)–(3), we are able to express the hyperfine driven nuclear spin dynamics in terms of the susceptibility and fluctuations of the electron spin density. Let us now explain how one

could calculate the necessary electronic correlation functions using linear response theory. For a given setup, we write down the full set of Bloch equations describing the evolution of the electronic density matrix, and solve for the steady state solution $\hat{\rho}^{(0)}$. The elements S^a are then simply found as the equilibrium expectation values $S^a = Av_0\langle\hat{S}^a(\mathbf{r}_n)\rangle_{\hat{\rho}^{(0)}} = \mathrm{Tr}\{\hat{S}^a(\mathbf{r}_n)\hat{\rho}^{(0)}\}$. Next, we add to the set of Bloch equations the first-order effect of a perturbation $\hat{H}' = \mathbf{\Lambda}\cdot\hat{\mathbf{S}}(\mathbf{r}_n)$, i.e., we add the terms $i[\hat{\rho}^{(0)},\hat{H}']_-$, and solve for the new steady state density matrix $\hat{\rho}^{(1)}$. The spin susceptibility χ^{ab} needed in Eq. (1) is then found as the part of $\langle\hat{S}^a(\mathbf{r}_n)\rangle_{\hat{\rho}^{(1)}} = \mathrm{Tr}\{\hat{S}^a(\mathbf{r}_n)\hat{\rho}^{(1)}\}$ which is linear in Λ^b. The fluctuations R^{ab} are found in a similar way, the terms to add to the Bloch equations then read $[\hat{\rho}^{(0)},\hat{H}']_+ - 2\mathrm{Tr}\{\hat{H}'\hat{\rho}^{(0)}\}\hat{\rho}^{(0)}$, where the last term removes the connected part of the electron spin correlators. We will illustrate the procedure below.

From this short outline, it is already clear that this approach has several advantages over a traditional FGR calculation: (i) For an FGR calculation, there should be an obvious basis to calculate all occupation probabilities in; here we can choose any basis which is most convenient for evaluating the steady state electronic density matrix. (ii) When calculating separate FGR transition rates, it should a priori be clear which quantization axis to choose for the nuclear spins; in our formalism, as soon as $\hat{\rho}^{(1)}$ is found for the two types of perturbations, all elements of χ and R can be read off immediately, yielding the dynamics for the full vector $\langle\hat{\mathbf{K}}_n\rangle$, not just the polarization along a single axis. (iii) For FGR to work, all levels involved in the hyperfine transitions (including possible virtual states in higher order processes) should be well separated in energy, i.e., the splittings should be larger than all decay rates and other possible incoherent processes present; in our method all incoherent processes are accounted for in the set of Bloch equations, we do not have to compare them with the other energy scales.

EXAMPLE APPLICATIONS

We will now illustrate the above with a calculation of the nuclear spin pumping in two example systems. First, we will consider nuclear spin pumping in a trivial toy model concerning a single quantum dot coupled to two leads. We will show how a smart implementation of FGR can produce an identical result in this case. Then we will investigate a more complicated setup, involving a double quantum dot, which cannot be dealt with in an FGR approach. We will show however how our formalism can be applied straightforwardly, yielding a general equation for the nuclear spin pumping in this setup.

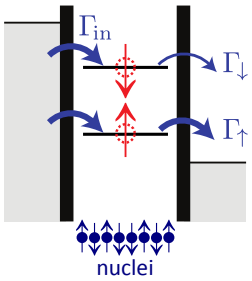

FIG. 1. Energy diagram for a trivial single quantum dot setup in which nuclear spin pumping is to be expected. The dot is coupled to two leads with different chemical potentials. One single-electron level lies within the bias window, which is split into two sublevels by the Zeeman energy (we assumed a negative g-factor). The tunnel rate from the left lead into the dot, Γ_{in} is large and equal for both spin directions. The outward tunnel rates $\Gamma_{\uparrow,\downarrow}$ are different for spin up and down.

Single dot: Fermi's golden rule with modifications

We consider a single quantum dot connected to two leads, as illustrated in Fig. 1. Due to the strong Coulomb repulsion, only one excess electron can occupy the dot. If we apply an external magnetic field $\mathbf{B}_{\mathrm{ext}} = B_{\mathrm{ext}}\hat{z}$, the energy levels for spin up and down in the dot are split by the electronic Zeeman energy. We take the tunnel rate into the dot Γ_{in} to be very fast and equal for both spin directions. The outward tunnel rates $\Gamma_{\uparrow,\downarrow}$ are different for the two spin directions (to provide a physical picture with this assumption: one could assume the right lead to be ferromagnetic). Since the electron occupying the dot will thus have on average a non-zero spin polarization along the z-direction, hyperfine interaction with the nuclear spins is expected to produce a non-zero nuclear spin polarization along this axis.

One usually assumes for simplicity that the electron spin is coupled equally strongly to all $N \sim 10^4$–10^6 nuclear spins. This then allows to rewrite the hyperfine Hamiltonian as $\hat{H}_{\mathrm{hf}}(t) = \sum_n A_n\hat{\mathbf{S}}(t)\cdot\hat{\mathbf{K}}_n(t)$, with the constant coefficients $A_n = A/N$ and $\hat{\mathbf{S}}$ being simply the electron spin operator (instead of spin density).

Let us calculate the nuclear spin pumping rate along the z-direction, $d\langle\hat{K}_n^z\rangle/dt$, for this system. Since all nuclear spins are coupled to $\hat{\mathbf{S}}$ with the same coefficient, $\langle\hat{K}_n^z\rangle$ for the n-th nucleus equals the ensemble averaged nuclear spin $\langle\hat{K}^z\rangle$. We only take into account a finite polarization in the z-direction, so we use the equation

$$\frac{d\langle\hat{K}^z\rangle}{dt} = \frac{1}{3}P^z - \frac{1}{2}Q^{zz}\langle\hat{K}^z\rangle, \tag{11}$$

with the functions $P^z = A_n^2K(K+1)(\chi^{xy}-\chi^{yx})$ and $Q^{zz} = A_n^2(R^{xx}+R^{yy})$. Note that χ and R contain now correlators of the electron spin, not electron spin density.

A finite nuclear polarization in the z-direction behaves on the time scale of the electronic dynamics as a static

contribution to the magnetic field experienced by the electrons. We incorporate this contribution into an effective field $B_{\rm eff} = B_{\rm ext} - A\langle\hat{K}^z\rangle$ (we assume a negative g-factor and a positive hyperfine coupling constant, as is the case in GaAs). The coherent time-evolution of the electronic 2×2 density matrix of this simple system is then given by $\partial_t\hat{\rho} = i[B_{\rm eff}\hat{S}^z, \hat{\rho}]_-$, where we express $B_{\rm eff}$ in terms of a frequency. To this we add the incoherent tunnel rates, yielding the set of equations

$$\begin{aligned}
\partial_t\rho_\uparrow &= -\Gamma_\uparrow\rho_\uparrow + \tfrac{1}{2}(\Gamma_\downarrow\rho_\downarrow + \Gamma_\uparrow\rho_\uparrow)\\
\partial_t\rho_\downarrow &= -\Gamma_\downarrow\rho_\downarrow + \tfrac{1}{2}(\Gamma_\downarrow\rho_\downarrow + \Gamma_\uparrow\rho_\uparrow)\\
\partial_t\rho_{\uparrow\downarrow} &= \quad iB_{\rm eff}\rho_{\uparrow\downarrow} - \tfrac{1}{2}\Gamma\rho_{\uparrow\downarrow}\\
\partial_t\rho_{\downarrow\uparrow} &= -iB_{\rm eff}\rho_{\downarrow\uparrow} - \tfrac{1}{2}\Gamma\rho_{\downarrow\uparrow},
\end{aligned} \tag{12}$$

where $\Gamma \equiv \Gamma_\uparrow + \Gamma_\downarrow$. These equations can be solved for the stationary situation $\partial_t\hat{\rho}^{(0)} = 0$, yielding $\rho^{(0)}_{\uparrow(\downarrow)} = \Gamma_{\downarrow(\uparrow)}/\Gamma$ and $\rho^{(0)}_{\uparrow\downarrow} = \rho^{(0)}_{\downarrow\uparrow} = 0$. We only need the response functions χ^{xy} and χ^{yx}, so we add the terms $i[\hat{\rho}^{(0)}, \Lambda_x\hat{S}_x + \Lambda_y\hat{S}_y]_-$ to Eq. (12) and solve for the new stationary solution $\hat{\rho}^{(1)}$. The correlator χ^{xy} is then simply given by the term in $\mathrm{Tr}\{\hat{S}^x\hat{\rho}^{(1)}\}$ linear in Λ_y, and in a similar way we find χ^{yx}. The results are combined to

$$\chi^{xy} - \chi^{yx} = \frac{\Gamma_\downarrow - \Gamma_\uparrow}{2B^2_{\rm eff} + \frac{1}{2}\Gamma^2}. \tag{13}$$

Now we evaluate R^{xx} and R^{yy}. To this end, we add the terms $[\hat{\rho}^{(0)}, \Lambda_x\hat{S}_x + \Lambda_y\hat{S}_y]_+ - 2\mathrm{Tr}\{(\Lambda_x\hat{S}_x + \Lambda_y\hat{S}_y)\hat{\rho}^{(0)}\}\hat{\rho}^{(0)}$ to Eq. (12), and then look for the linear responses in $\hat{S}^x$ and $\hat{S}^y$, yielding

$$R^{xx} + R^{yy} = \frac{\Gamma}{2B^2_{\rm eff} + \frac{1}{2}\Gamma^2}. \tag{14}$$

Combining all together using Eq. (11), we thus find

$$\frac{d\langle\hat{K}^z\rangle}{dt} = A_n^2\frac{\Gamma_\downarrow(\frac{1}{2} - \langle\hat{K}^z\rangle) - \Gamma_\uparrow(\frac{1}{2} + \langle\hat{K}^z\rangle)}{4(B_{\rm ext} - A\langle\hat{K}^z\rangle)^2 + \Gamma^2}, \tag{15}$$

where for simplicity we assumed $K = 1/2$.

Let us compare the result (15) with the pumping rate we get using an FGR approach. We focus on transitions between the two states $|\uparrow\Downarrow_n\rangle$ and $|\downarrow\Uparrow_n\rangle$, where the double arrow indicates the nuclear spin along the z-axis. Neglecting the nuclear Zeeman energy, the two states are separated by the energy $\hbar B_{\rm eff}$, and both are broadened by their respective electronic decay rate $\Gamma_{\uparrow(\downarrow)}$. Hyperfine induced transitions from $|\uparrow\Downarrow_n\rangle$ to $|\downarrow\Uparrow_n\rangle$ contribute *positively* to $d\langle\hat{K}^z_n\rangle/dt$, and their rate is given by

$$\Gamma_+ = 2\pi|\,\langle\downarrow\Uparrow_n|\tfrac{1}{2}A_n\hat{S}^-\hat{K}^+_n|\uparrow\Downarrow_n\rangle\,|^2 p_i\,\mathcal{D}, \tag{16}$$

the square of the relevant matrix element, multiplied by the chance p_i of finding the system initially in $|\uparrow\Downarrow_n\rangle$ and the effective 'density of states' for this transition $\mathcal{D}$.

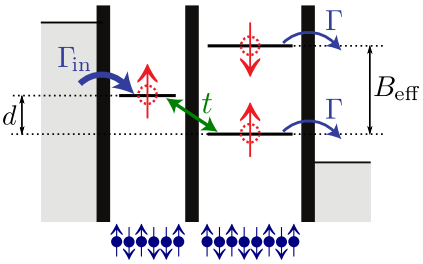

FIG. 2. Energy diagram of a more complicated setup. Two quantum dots are coupled to each other and to two leads with different chemical potentials. The two dots have different effective g-factors, so that the application of an external magnetic field yields different Zeeman energies in the dots. The double dot is tuned to the $(0,0)\to(1,0)\to(0,1)\to(0,0)$ transport regime. The outward tunneling rates Γ are equal and much smaller than the inward rate $\Gamma_{\rm in}$.

For the occupation probability p_i, we take $p_i = (\frac{1}{2} - \langle\hat{K}^z\rangle)\times\rho^{(0)}_\uparrow$. In this simple case, it is possible to incorporate all incoherent dynamics (the two decay rates) into a smartly chosen $\mathcal{D}$: We take the sum of the level broadenings of the two electronic levels $|\uparrow\rangle$ and $|\downarrow\rangle$. Assuming a Lorentzian shape for these broadenings, we thus use

$$\mathcal{D} = \frac{1}{\pi}\frac{\frac{1}{2}\Gamma}{B^2_{\rm eff} + \frac{1}{4}\Gamma^2}, \tag{17}$$

so that we can write

$$\Gamma_+ = A_n^2\frac{\Gamma_\downarrow(\frac{1}{2} - \langle\hat{K}^z\rangle)}{4B^2_{\rm eff} + \Gamma^2}. \tag{18}$$

We derive similarly an equation for Γ_- and then combine the two to write for the net nuclear spin pumping rate

$$\frac{d\langle\hat{K}^z\rangle}{dt} = A_n^2\frac{\Gamma_\downarrow(\frac{1}{2} - \langle\hat{K}^z\rangle) - \Gamma_\uparrow(\frac{1}{2} + \langle\hat{K}^z\rangle)}{4(B_{\rm ext} - A\langle\hat{K}^z\rangle)^2 + \Gamma^2}. \tag{19}$$

We see that this result coincides with Eq. (15). The fact that the FGR approach in this case also works for small energy splittings, i.e., in the regime $B_{\rm eff} \lesssim \Gamma$, is due to the fact that all incoherent effects in this model can be incorporated relatively simply into $\mathcal{D}$.

Double dot: Beyond Fermi's golden rule

Let us now illustrate what can happen when the setup becomes more complicated. We consider a double quantum dot connected to two leads with different chemical potentials, as depicted in Fig. 2. The dots are tuned such that an electronic transport cycle involves the charge states $(0,0)\to(1,0)\to(0,1)\to(0,0)$, where (n,m) denotes a state with $n(m)$ excess electrons on the left(right) dot. The electronic levels in the two dots are both split by a Zeeman energy in a spin up and spin down level. We

assume that the splitting in the left dot is larger than in the right dot (e.g. due to a size-related difference in effective g-factors), and that the $(1,0)$ spin down level has a too high energy to play a role. The $(1,0)$ and $(0,1)$ spin up levels are detuned by an energy d and coupled to each other with a tunnel coupling t. We again assume the outward tunneling rates to be much slower than the inward, $\Gamma \ll \Gamma_{\rm in}$, so that the system effectively will never occupy the $(0,0)$ charge state.

We will focus on nuclear spin pumping in the right dot. A non-zero nuclear spin pumping rate is expected along the direction of the magnetic field since the average local electron spin polarization along this direction is finite. Denoting the level in the left dot by $|L\rangle$ and the two levels in the right dot by $|\uparrow\rangle$ and $|\downarrow\rangle$, the Hamiltonian for the three-level system reads

$$\hat{H}_3 = d\,|L\rangle\langle L| + B_{\rm eff}\,|\downarrow\rangle\langle\downarrow| + t\,|L\rangle\langle\downarrow| + t\,|\downarrow\rangle\langle L|\,, \quad (20)$$

where the feedback of the nuclear spins on the electron spin dynamics again is incorporated into the effective magnetic field $B_{\rm eff} = B_{\rm ext} - A\langle\hat{K}^z\rangle$. Using this Hamiltonian and adding the effect of the incoherent decay rate Γ, we can write down the equations of motion for $\hat{\rho}$, and solve for $\hat{\rho}^{(0)}$, yielding

$$\begin{aligned} \rho_L^{(0)} &= \frac{4d^2+4t^2+\Gamma^2}{4d^2+8t^2+\Gamma^2}, \quad \rho_\uparrow^{(0)} = \frac{4t^2}{4d^2+8t^2+\Gamma^2}, \\ \rho_{L\uparrow}^{(0)} &= (\rho_{\uparrow L}^{(0)})^* = \frac{4dt+2it\Gamma}{4d^2+8t^2+\Gamma^2}, \end{aligned} \quad (21)$$

and zero for the other five elements. We then proceed as we did above, and add consecutively the perturbations $i[\hat{\rho}^{(0)},\hat{H}']_-$ and $[\hat{\rho}^{(0)},\hat{H}']_+ - 2{\rm Tr}\{\hat{H}'\hat{\rho}^{(0)}\}\hat{\rho}^{(0)}$ to the equations of motion. We solve in both cases for the new steady-state solution $\hat{\rho}^{(1)}$ and extract the linear responses needed. Combining all together, we find

$$\frac{d\langle\hat{K}^z\rangle}{dt} = \frac{4A_n^2t^2\Gamma(B_{\rm eff}^2+2t^2+\Gamma^2)(\frac{1}{2}-\langle\hat{K}^z\rangle)}{(4d^2+8t^2+\Gamma^2)[4(B_{\rm eff}d-B_{\rm eff}^2+t^2)^2+(5B_{\rm eff}^2-8B_{\rm eff}d+4d^2+4t^2)\Gamma^2+\Gamma^4]}, \quad (22)$$

where $B_{\rm eff} = B_{\rm ext} - A\langle\hat{K}^z\rangle$. This equation gives the nuclear spin pumping in the right dot along the z-axis, without any restriction imposed on the relative magnitude of the parameters $B_{\rm eff}$, d, t, and Γ.

Let us now try to evaluate the same pumping using an FGR approach. The most transparent electronic basis to use is $\{|L\rangle, |\uparrow\rangle, |\downarrow\rangle\}$ since in this basis we have well-defined incoherent in- and outward tunneling rates. We see however that the stationary density matrix in this basis (21) has off-diagonal elements. In order to proceed, we have to make several assumptions. Let us consider the limit of $\Gamma \gg t$. In this case the off-diagonal elements of $\hat{\rho}^{(0)}$ can be neglected, and we can approximate $\hat{\rho}^{(0)} \approx |L\rangle\langle L|$. We further assume that we have three well-separated levels, i.e., $B_{\rm eff}, d \gg \Gamma$. In this limit the nuclear spin pumping is resulting from a second order transition: (i) tunneling from the initial state $|L\Uparrow_n\rangle$ to the virtual state $|\downarrow\Uparrow_n\rangle$, and then (ii) an electron-nuclear spin flip-flop from $|\downarrow\Uparrow_n\rangle$ to the final state $|\uparrow\Downarrow_n\rangle$. We use a standard second order Fermi golden rule to write for the pumping rate

$$\frac{d\langle\hat{K}^z\rangle}{dt} = 2\pi\frac{|\langle\downarrow\Uparrow_n|\frac{1}{2}A_n\hat{S}^-\hat{K}_n^+|\uparrow\Downarrow_n\rangle\langle\uparrow\Downarrow_n|\hat{H}_T|L\Downarrow_n\rangle|^2}{(E_{\downarrow\Uparrow_n}-E_{L\Uparrow_n})^2}p_{L\Uparrow_n}\mathcal{D}, \quad (23)$$

where $\hat{H}_T = t\,|L\rangle\langle\downarrow| + t\,|\downarrow\rangle\langle L|$ is the tunneling part of the Hamiltonian $\hat{H}_3$. The occupation probability of the initial state is $p_{L\Uparrow_n} = \frac{1}{2} - \langle\hat{K}^z\rangle$. The 'density' $\mathcal{D}$ for this transition is mainly set by the level broadening of the final state, so $\mathcal{D} = \frac{2}{\pi}\Gamma/[4(B_{\rm eff}-d)^2+\Gamma^2]$. This yields

$$\frac{d\langle\hat{K}^z\rangle}{dt} = \frac{A_n^2t^2\Gamma(\frac{1}{2}-\langle\hat{K}^z\rangle)}{d^2[4(B_{\rm eff}-d)^2+\Gamma^2]}, \quad (24)$$

which coincides with (22) in the limit $B_{\rm eff}, d \gg \Gamma \gg t$. (Note that $B_{\rm eff} - d \gg \Gamma$ is not a necessary condition.)

There are other limits in which considering the right rates to calculate, combined with a good approximation for $\hat{\rho}^{(0)}$ and a smart choice for $\mathcal{D}$, can result in the right pumping rate. In general however, it is not trivial to understand how to incorporate all incoherent dynamics into $\mathcal{D}$. Even if one would simply diagonalize the stationary density matrix $\hat{\rho}^{(0)}$ and try to evaluate all hyperfine transitions in the new basis, one would have to transform the incoherent processes in correct effective densities of state. In the approach using the electron spin susceptibility and fluctuations, this is all done on the fly and its validity is not restricted to certain limits of the parameter space.

Let us emphasize here that the fact that a correctly adapted FGR approach did work in our single dot example but not in the double dot example, is only due to the complexity of the coherent and incoherent electron spin dynamics in the two cases. The success of the FGR approach is not inherently connected to the number of quantum dots in a setup: Also for more complicated single dot systems, the FGR approach might break down.

The two examples given above both consider localized electron spins. Our approach however, works in princi-

ple equally well for systems with delocalized electrons. In this case, as explained above, the susceptibility and fluctuations needed are correlators of electron spin *density* instead of simply electron spin. This however does not change the complexity of the formalism: One only has to find the right set of (Bloch) equations to describe the electron spin density.

CONCLUSIONS

To conclude, we presented a straightforward approach to evaluate the nuclear spin dynamics driven by hyperfine interaction with non-equilibrium electron spins. To describe the dynamics up to second order in the hyperfine coupling, it suffices to evaluate the susceptibility and fluctuations of the electron spin. This approach does not rely on a separation of electronic energy scales or the specific choice of electronic basis states, thereby overcoming practical problems which may arise when using a more traditional formalism based on rate equations.

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[22] Similarly, the electronic correlation functions in (3) to leading order only involve connected terms: χ^{ab}_{nm} per definition, and R^{ab} while the unconnected terms yield only corrections to the first order equations of motion.