Stochastic dynamics and control of a driven nonlinear spin chain: the role of Arnold diffusion

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# Stochastic dynamics and control of a driven nonlinear spin chain: the role of Arnold diffusion 

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#### Abstract

We study a chain of nonlinear interacting spins driven by a static and a time-dependent magnetic field. The aim is to identify the conditions for the locally and temporally controlled spin switching. Analytical and full numerical calculations show the possibility of stochastic control if the underlying semiclassical dynamics is chaotic. This is achievable by tuning the external field parameters according to the method described in this paper. We show analytically for a finite spin chain that Arnold diffusion is the underlying mechanism for the present stochastic control. Quantum mechanically we consider the regime where the classical dynamics is regular or chaotic. For the latter we utilize the random matrix theory. The efficiency and the stability of the non-equilibrium quantum spin states are quantified by the time dependence of the Bargmann angle related to the geometric phases of the states.


## 1. Introduction

Advances in nanoscale fabrication of magnetic materials down to a finite chain of individual magnetic atoms [1] triggered a number of studies on the ground state magnetic properties of finite, interacting spin chains [2]. For accessing the nonequilibrium states, in a linear chain one conventionally rotates the spins by applying a static magnetic field $\mathbf{H}_{0}$ and a variable magnetic field $\mathbf{h}(t)$ along a direction perpendicular to $\mathbf{H}_{0}$ [3]. The spins affected by the fields are then deflected by an angle $\theta=\omega_{1} \tau$ which can be desirably varied by changing the duration $\tau$ of the field $\mathbf{h}(t)$. Here $\omega_{1}$ is the amplitude of $\mathbf{h}(t)$ in frequency units $\omega_{1}=h_{0} \gamma$ ( $\gamma$ is the gyromagnetic ratio). For this scheme to be viable $\omega_{1}$ has to be in resonance with the system's precessional frequency. In this paper we consider the spin deflection in the different situation of a nonlinear chain of interacting spins [4, 5] in which case the precessional frequency is dynamical and changes with the oscillation amplitude [6, 7]. Hence, a control strategy [8, 9] as in the linear chain case entails the use of chirped fields. Here we inspect a different route to spin control by exploiting the stochastic nature of the spin dynamics when appropriate fields are employed. This we show analytically in a first step. The advantage is that no special frequency tuning is used and, more
importantly, the spin may be quasi-stable at the deflected (nonequilibrium) angles when the field $\mathbf{h}(t)$ is off, which might be of interest for quantum information applications [10-17]. The disadvantage is the limited control of the switching time. Full numerical simulations confirm our analytical predictions: tuning the external fields such that the underlying classical spin dynamics is chaotic, stochastic switching occurs and a long-time quasi-stabilization, i.e. a dynamical freezing (DF) of the deflected states, is possible. Small fields cause only small fluctuations around the equilibrium state. For very strong fields, effects of magnetic anisotropy and exchange become subsidiary and hence the dynamics turn regular and no deflection with subsequent freezing is possible.

For a finite spin chain we uncover analytically that our stochastic control (SC) scheme is governed by Arnold diffusion and give an analytical expression for the Arnold diffusion coefficient that in turn determines the timescale for SC.

To inspect the influence of the quantum nature of the spins on our (classical) predictions we considered both the regular and the chaotic classical regimes and evaluated the so-called Bargmann angle which is a measure of the quantum distance between states in the Hilbert space and can be used to signal DF $[8,9]$. Using random matrix theory we prove indeed that SC and DF are possible at the driving field values that follow from our classical analysis.


Figure 1. A schematics of the interacting spin chain. The magnetic anisotropy field sets the $z$ direction. Two magnetic fields are applied: a static $\left(H_{0 z}\right)$ one along the $z$ axis and a time-dependent field $\left(H_{0 x}(t)\right)$ along the $x$ axis.
(This figure is in colour only in the electronic version)

## 2. Equation of motion

### 2.1. Liouville equation for the spin chain

Similar to the case studied in $[4,5]$ we consider a system that can be modelled by a chain of $N$ interacting (with coupling constant $J$ ) spin variables localized at sites $j$ and having a uniaxial anisotropy with the anisotropy constant $\beta$. Possible sources of the anisotropy field are discussed in [1]. Here we mention that the inclusion of a finite anisotropy is essential for the existence of a finite-temperature long-range order in the (infinitely long) chain. Further important consequences of the magnetic anisotropy for the phenomena discussed in this work are detailed below. The direction of the anisotropy field defines the $z$ axis. A static $\left(H_{0 z}\right)$ and a time-dependent $\left(H_{0 x}(t)\right)$ magnetic field are applied along the $z$ and $x$ axes, respectively (cf figure 1). $H_{0 x}(t)$ consists of $N_{k}$ periodic pulses, i.e.

$$
\begin{equation*}
H_{0 x}(t)=\varepsilon T \sum_{k}^{N_{k}} \delta(t-k T) \tag{1}
\end{equation*}
$$

where $T$ is the period and $\varepsilon T$ is the field strength. As demonstrated explicitly [18] (for the classical spin dynamics) the shape (1) of the field mimics well the action of a finitewidth pulse as long as the pulse duration is smaller than the field-free precessional period of the spins. The time integral over the field amplitude of the finite-width pulse sets the variable $\varepsilon$ [18].

From the Hamilton operator $[4,5]$
$\hat{H}=\sum_{j=1}^{N-1} J \hat{s}_{j z} \hat{s}_{j+1, z}+H_{0 x} \sum_{j=1}^{N} \hat{s}_{j x}+H_{0 z} \sum_{j=1}^{N} \hat{s}_{j z}+\beta \sum_{j=1}^{N} \hat{s}_{j z}^{2}$
we find the spin equation of motion (EOM) to be

$$
\begin{gather*}
\partial_{t} \hat{\mathbf{s}}_{j}=\mathbf{h}_{j} \times \hat{\mathbf{s}}_{j}, \\
\mathbf{h}_{j}=\left(H_{0 x}, 0, J\left[\hat{s}_{j+1 z}+\hat{s}_{j-1 z}\right]+H_{0 z}+2 \beta \hat{s}_{j z}\right) \tag{3}
\end{gather*}
$$

For large spins $s_{j}$ with $\hbar s_{j}\left(s_{j}+1\right)=\left\langle\hat{\mathbf{s}}_{j}\right\rangle$ and $\left[\hat{H}, \hat{\mathbf{s}}_{i}^{2}\right]=0$ we shift variables as (cf figure $1, s_{i}^{2}=1$ )

$$
\begin{gather*}
s_{i x}=s_{i \perp} \cos \varphi_{i} ; \quad s_{i y}=s_{i \perp} \sin \varphi_{i}, \\
s_{i, \perp}=\sqrt{1-s_{i, z}^{2}} . \tag{4}
\end{gather*}
$$

The EOM for two spins $N=2$ in terms of the canonical action variables $s_{1 z}, s_{2 z}$ and their conjugate angles $\varphi_{1}, \varphi_{2}$ is

$$
\begin{align*}
& \dot{s}_{i z}=-\varepsilon \frac{\partial V}{\partial \varphi_{i}}, \quad \dot{\varphi}_{i}=\omega_{i}\left(s_{j z}\right)+\varepsilon \frac{\partial V}{\partial s_{i z}} \\
& \omega_{i}\left(s_{j z}\right)=J s_{j z}+2 \beta \hat{s}_{i z}+H_{0 z}, \quad i, j \in\{1,2\} \\
& V:=\sum_{i=1}^{N} V_{i}\left(s_{i z}, \varphi_{i}\right)=\sum_{i=1}^{N} V_{0 i}\left(s_{i z}, \varphi_{i}\right)  \tag{5}\\
& \quad \times T \sum_{k=-\infty}^{+\infty} \delta(t-k T)=\sum_{i=1}^{N} s_{i \perp} \cos \varphi_{i} \\
& \quad \times T \sum_{k=-\infty}^{+\infty} \delta(t-k T)
\end{align*}
$$

For $V \neq 0$ the variables of actions are adiabatic invariants and hence are slow with respect to the angle's typical timescale [6]. The idea now is to identify the regime of classical chaotic dynamics which we will do below. In this regime one may adopt a kinetic approach based on the Liouville equation $[19,20]$ for the two-particle distribution function $f\left(t, s_{1 z}, \varphi_{1}, s_{2 z}, \varphi_{2}\right)$, i.e.
$\mathrm{i} \frac{\partial f}{\partial t}=\left(\hat{L}_{0}+\varepsilon \hat{L}_{1}\right) f$,

$$
\begin{align*}
\hat{L}_{0}= & -i \omega_{1}\left(s_{2 z}\right) \frac{\partial}{\partial \varphi_{1}}-i \omega_{2}\left(s_{1 z}\right) \frac{\partial}{\partial \varphi_{2}} \\
\hat{L}_{1} & =-i\left(\frac{\partial V}{\partial s_{1 z}} \frac{\partial}{\partial \varphi_{1}}-\frac{\partial V}{\partial \varphi_{1}} \frac{\partial}{\partial s_{1 z}}\right)  \tag{6}\\
& -i\left(\frac{\partial V}{\partial s_{2 z}} \frac{\partial}{\partial \varphi_{2}}-\frac{\partial V}{\partial \varphi_{2}} \frac{\partial}{\partial s_{2 z}}\right) .
\end{align*}
$$

Equation (6) is of key importance for this study. Below we use the random phase approximation and some mathematical techniques to derive from equation (6) the Fokker-Planck equation which allows us to explore some chaotic features of the spin dynamics.

### 2.2. Fokker-Planck formulation and the onset of the chaotic regime

Expressing $f\left(s_{1 z}, \varphi_{1}, s_{2 z}, \varphi_{2}\right)$ as a Fourier series over $\varphi_{1}$ and $\varphi_{2}$ we find
$f\left(s_{1 z}, \varphi_{1}, s_{2 z}, \varphi_{2}\right)=\frac{1}{(2 \pi)^{2}} \sum_{m, n} \bar{f}_{n, m}\left(s_{1 z}, s_{2 z}\right) \mathrm{e}^{\mathrm{i} n \varphi_{1}} \mathrm{e}^{\mathrm{i} m \varphi_{2}}$,
$\bar{f}_{n, m}\left(s_{1 z}, s_{2 z}\right)=f_{n, m}\left(s_{1 z}, s_{2 z}\right)$
$\times \exp \left[-\mathrm{i} n \int_{0}^{t} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}\right] \exp \left[-\mathrm{i} m \int_{0}^{t} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}\right]$,
$\omega_{1}\left(t^{\prime}\right)=\omega_{1}\left(s_{2 z}\left(t^{\prime}\right)\right), \quad \omega_{2}\left(t^{\prime}\right)=\omega_{2}\left(s_{1 z}\left(t^{\prime}\right)\right)$.
Hence, solution of the Liouville equation is cast formally as (the symbol $\left\langle n^{\prime}, m^{\prime}\right| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle$ means the average over fast oscillating variables)

$$
\begin{align*}
& f_{n^{\prime}, m^{\prime}}\left(s_{1 z}, s_{2 z}, t\right)=f_{n^{\prime}, m^{\prime}}\left(s_{1 z}, s_{2 z}, 0\right) \\
& \quad-\mathrm{i} \varepsilon \sum_{n, m} \int_{0}^{t} \mathrm{~d} t_{1} \mathrm{e}^{\mathrm{i}\left(n^{\prime}-n\right) \int_{0}^{t_{1}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \mathrm{e}^{\mathrm{i}\left(m^{\prime}-m\right) \int_{0}^{t_{1}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \\
& \quad \times\left\langle n^{\prime} m^{\prime}\right| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle f_{n, m}\left(s_{1 z}, s_{2 z}, t_{1}\right) \tag{8}
\end{align*}
$$

If the interaction energy with the variable field is small with respect to the other terms in equation (2) we can expand $f$ in terms of the field strength $\varepsilon$ and account for leading terms only. The zero-order component has the form

$$
\begin{align*}
& f_{0,0}\left(s_{1 z}, s_{2 z}, t\right)=f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right) \\
& \quad-\mathrm{i} \varepsilon \sum_{n, m} \int_{0}^{t} \mathrm{~d} t_{1} \mathrm{e}^{-\mathrm{i} n \int_{0}^{t_{1}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \mathrm{e}^{-\mathrm{i} m \int_{0}^{t_{1}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \\
& \quad \times\langle 0,0| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle f_{n, m}\left(s_{1 z}, s_{2 z}, 0\right) \\
& \quad+(-\mathrm{i} \varepsilon)^{2} \sum_{n, m} \int_{0}^{t} \mathrm{~d} t_{1} \int_{0}^{t_{1}} \mathrm{~d} t_{2} \mathrm{e}^{\mathrm{i} n \int_{t_{1}}^{t_{1}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \mathrm{e}^{\mathrm{i} m \int_{t_{1}}^{t_{2}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \\
& \quad \times\langle 0,0| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle\langle n, m| \hat{L}_{1}\left(t_{2}\right)|0,0\rangle \\
& \quad \times f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right) \tag{9}
\end{align*}
$$

Now we write $\hat{L}_{1}$ as a Fourier series taking the relevant frequency $\Omega=\frac{2 \pi}{T}$ into account, $\hat{L}_{1}(t)=$ $\sum_{p} L_{1, p} \exp (\mathrm{i} p \Omega t), L_{1,-p}=L_{1, p}^{*}$. Inserting into (9) we find

$$
f_{0,0}\left(s_{1 z}, s_{2 z}, t\right)=f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right)
$$

$$
-\mathrm{i} \varepsilon \sum_{n, m} \int_{0}^{t} \mathrm{~d} t_{1} \mathrm{e}^{-\mathrm{i} n \int_{0}^{t_{1}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \mathrm{e}^{-\mathrm{i} m \int_{0}^{t_{1}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}}
$$

$$
\times\langle 0,0| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle f_{n, m}\left(s_{1 z}, s_{2 z}, 0\right)
$$

$$
+(-\mathrm{i} \varepsilon)^{2} \sum_{n, m, p} \int_{0}^{t} \mathrm{~d} t_{1} \int_{0}^{t_{1}} \mathrm{~d} t_{2} \mathrm{e}^{\mathrm{i} n \int_{t_{1}}^{t_{1}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}} \mathrm{e}^{\mathrm{i} m \int_{t_{1}}^{t_{2}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime}}
$$

$$
\times\langle 0,0| \hat{L}_{1, p}\left(t_{1}\right)|n, m\rangle\langle n, m| \hat{L}_{1,-p}\left(t_{2}\right)|0,0\rangle
$$

$$
\begin{equation*}
\times \mathrm{e}^{\mathrm{i} p \Omega\left(t_{1}-t_{2}\right)} f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right) \tag{10}
\end{equation*}
$$

Introducing the notations

$$
\begin{equation*}
\psi_{1}\left(t_{1}, t_{2}\right)=\int_{t_{1}}^{t_{2}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime}, \quad \psi_{2}\left(t_{1}, t_{2}\right)=\int_{t_{1}}^{t_{2}} \omega_{2}\left(t^{\prime}\right) \mathrm{d} t^{\prime} \tag{11}
\end{equation*}
$$

we write

$$
\begin{align*}
& f_{0,0}\left(s_{1 z}, s_{2 z}, t\right)=f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right) \\
& \quad-\mathrm{i} \varepsilon \sum_{n, m} \int_{0}^{t} \mathrm{~d} t_{1} \mathrm{e}^{-\mathrm{i} n \psi_{1}\left(t_{1}, 0\right)} \mathrm{e}^{-\mathrm{i} m \psi_{2}\left(t_{1}, 0\right)} \\
& \quad \times\langle 0,0| \hat{L}_{1}\left(t_{1}\right)|n, m\rangle f_{n, m}\left(s_{1 z}, s_{2 z}, 0\right) \\
& \quad+(-\mathrm{i} \varepsilon)^{2} \sum_{n, m, p} \int_{0}^{t} \mathrm{~d} t_{1} \int_{0}^{t_{1}} \mathrm{~d} t_{2} \mathrm{e}^{-\mathrm{i} n \psi_{1}\left(t_{1}, t_{2}\right)} \mathrm{e}^{-\mathrm{i} m \psi_{2}\left(t_{1}, t_{2}\right)} \\
& \quad \times\langle 0,0| \hat{L}_{1, p}|n, m\rangle\langle n, m| \hat{L}_{1,-p}|0,0\rangle \\
& \quad \times \mathrm{e}^{\mathrm{i} p \Omega\left(t_{1}-t_{2}\right)} f_{0,0}\left(s_{1 z}, s_{2 z}, 0\right) \tag{12}
\end{align*}
$$

Averaging over the initial phases, i.e. $F\left(s_{1 z}, s_{2 z}, t\right)=$ $\left\langle\left\langle f_{0,0}\left(s_{1 z}, s_{2 z}, t\right)\right\rangle\right\rangle$, and using the random phase approximation $\psi_{1}\left(t_{1}, t_{2}\right)=\int_{t_{1}}^{t_{2}} \omega_{1}\left(t^{\prime}\right) \mathrm{d} t^{\prime} \approx \varphi_{1}\left(t_{2}\right)-\varphi_{1}\left(t_{2}\right)$, we end up with

$$
\begin{align*}
& \left\langle\left\langle\exp \left[\operatorname{in} \psi_{1,2}\left(t_{2}, t_{1}\right)\right]\right\rangle\right\rangle=\exp \left(-\left(t_{1}-t_{2}\right) / \tau_{\mathrm{c}}\right) \\
& \quad \times \exp \left(-\mathrm{i} n \omega_{1,2}\left(t_{1}-t_{2}\right)\right) . \tag{13}
\end{align*}
$$

Here $\tau_{\mathrm{c}}$ is the correlation time of the random phase. Taking equation (12) into account we deduce then for the averaged two-particle distribution function $F(t)$ the dynamical equation
(up to second order in the field strength $\varepsilon$ )

$$
\begin{align*}
\frac{\partial F}{\partial t} & =-\mathrm{i} \varepsilon \mathrm{e}^{-\frac{2 t}{\tau_{c}}} \sum_{n, m} \mathrm{e}^{-\mathrm{i}\left(n \omega_{1}+m \omega_{2}\right) t} \\
& \times\langle 0,0| \hat{L}_{1}(t)|n, m\rangle f_{n, m}\left(s_{1 z}, s_{2 z}, 0\right) \\
& -\varepsilon^{2} \frac{\partial}{\partial t} \sum_{m, n, p} \int_{0}^{t} \mathrm{~d} t_{1} \int_{0}^{t_{1}} \mathrm{~d} t_{2} \mathrm{e}^{-\frac{2\left(t_{1}-t_{2}\right)}{\tau_{c}}} \mathrm{e}^{-\mathrm{i} n \omega_{1}\left(t_{1}-t_{2}\right)} \\
& \times \mathrm{e}^{-\mathrm{i} m \omega_{2}\left(t_{1}-t_{2}\right)} \mathrm{e}^{\mathrm{i} p \Omega\left(t_{1}-t_{2}\right)} \\
& \times\langle 0,0| L_{1, p}|n, m\rangle\langle m, n| L_{1,-p}|0,0\rangle F\left(s_{1 z}, s_{2 z}, t\right) \tag{14}
\end{align*}
$$

The long-time behaviour $\left(t \gg \tau_{\mathrm{c}}\right)$ is retrieved by shifting to the new variables $\tau=t_{1}-t_{2}, t_{1}=t_{1}$ in (14) and integrating over $\tau$ which yields

$$
\begin{align*}
\frac{\partial F}{\partial t} & =-\varepsilon^{2} \sum_{n, m, p} \frac{1}{\frac{2}{\tau_{\mathrm{c}}}+\mathrm{i}\left(n \omega_{1}+m \omega_{2}-p \Omega\right)} \\
& \times\langle 0,0| L_{1 p}|n, m\rangle\langle m, n| L_{1,-p}|0,0\rangle F\left(s_{1 z}, s_{2 z}, t\right) \tag{15}
\end{align*}
$$

For further progress explicit expressions for the matrix elements $\langle 0,0| L_{1, p}|n, m\rangle\langle m, n| L_{1,-p}|0,0\rangle$ are needed. Following the standard procedure outlined in [21] we find after some lengthy steps the following Fokker-Planck equation for $F\left(s_{1 z}, s_{2 z}, t\right)$ :
$\frac{\partial F\left(s_{1 z}, s_{2 z}, t\right)}{\partial t}$
$=D\left(\frac{\partial}{\partial s_{1 z}}\left(1-s_{1 z}^{2}\right) \frac{\partial F}{\partial s_{1 z}}+\frac{\partial}{\partial s_{2 z}}\left(1-s_{2 z}^{2}\right) \frac{\partial F}{\partial s_{2 z}}\right)$,
$D=\frac{\varepsilon^{2} \pi}{2 \Omega}=\frac{\varepsilon^{2} T}{4}$.
Making the ansatz $F\left(s_{1 z}, s_{2 z}\right)=F_{1}\left(s_{1 z}\right) F_{2}\left(s_{2 z}\right)$ the average values of the spin projections $\bar{s}_{j z}$ are determined from

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t} \bar{s}_{j z}=\int_{-1}^{+1} s_{j z} \frac{\partial F_{j}}{\partial t} \mathrm{~d} s_{j z}=D \int_{-1}^{+1} s_{1 z} \frac{\partial}{\partial s_{j z}}\left(1-s_{j z}^{2}\right) \\
& \quad \times \frac{\partial F_{j}\left(s_{j z}\right)}{\partial s_{j z}}=-2 D \bar{s}_{j z} ; \quad j=1,2  \tag{17}\\
& \bar{s}_{1,2 z}= \\
& s_{1,2 z}(0) \mathrm{e}^{-2 D t}
\end{align*}
$$

As discussed in [22] (for the case without an anisotropy field), essential for the validity of this diffusion type dynamics is that the underlying classical dynamics is chaotic in which case the above derivations are justified.

The stroboscopic evolution of the spin variables before $\left(t_{0}-\tau\right)$ and after $\left(t_{0}+\tau\right)$ applying the field pulses at $t=t_{0}$ is expressed as [22]
$s_{i z, n+1}=s_{i z, n}+\varepsilon T \sqrt{1-s_{i z, n}^{2}} \sin \varphi_{i, n}$,
$\varphi_{i, n+1}=\varphi_{i, n}+\left(J s_{j z, n+1}+2 \beta s_{i z}-H_{0 z}\right) T$
$-\varepsilon T \frac{s_{i z, n}}{\sqrt{1-s_{i z, n}^{2}}} \cos \varphi_{i, n}$.
The stability of the trajectories is deduced from the Jacobian matrix [23] which also sets the condition for the chaotic regime


Figure 2. The time evolution of $s_{1 z}$ and $s_{2 z}$ for the following parameters of the system: $J=0.2,2 \beta=0,1, H_{0 z}=0.2, \varepsilon=0.057$ and $\Omega=100$. $\frac{\tau_{0}}{T}=0.1, s_{1 z}(0)=0.8, \varphi_{1}(0)=0, s_{2 z}(0)=-0.8$, $\varphi_{2}(0)=0, T=\frac{2 \pi}{\Omega}$ and $K_{0}=K^{\prime}=0.45 \times 10^{-4}$.
as $\left(\langle\cdots\rangle_{t}\right.$ means time average)
$\left|\lambda_{i}\right|>1, \quad K>0$,
$\lambda_{1,2}=\frac{(2+K) \pm \sqrt{(K+2)^{2}-4}}{2}$,
$\lambda_{3,4}=\frac{(2-K) \pm \sqrt{(K-2)^{2}-4}}{2}$,
$K=K_{0} \sqrt{(\beta / J)^{2}+\left(1-2(\beta / J)^{2}\right) s_{1 \perp} s_{2 \perp}\left\langle\cos \varphi_{1} \cos \varphi_{2}\right\rangle_{t}}$,

$$
\begin{equation*}
K_{0}=\varepsilon T^{2} J \tag{20}
\end{equation*}
$$

Hence we can tune to the chaotic regime by varying the external field parameters, the constant of anisotropy $\beta$ and the coupling constant $J$ between adjacent spins. For evaluating averages of the form $\langle\cdots\rangle_{t}$ averages over time correlation functions of the random phases $\left\langle\cos \varphi_{1} \cos \varphi_{2}\right\rangle$ should be considered. For the correlation term we proceed as follows: when deriving the diffusion equation we assumed that correlation times of random phases are small with respect to the diffusion scale $\tau_{\mathrm{c}} \ll \frac{2 D}{\pi \varepsilon}=1 / \Omega$. Taking into account that in this timescale values $s_{1,2 \perp}$ are slow in time, after averaging the correlation functions over the time interval $\Delta t \in(0,1 / \Omega)$ we obtain $K=K_{0} \sqrt{(\beta / J)^{2}+\left(1-2(\beta / J)^{2}\right) \frac{\varepsilon \pi}{2 D} s_{1 \perp} s_{2 \perp} \tau_{\mathrm{c}}(J)}$.

### 2.3. Discussions and numerical results

Having discussed the analytical structure of the spin dynamics we compare the analytical predictions with full numerical simulations of the problem. Note that our system is such that the conditions of chaotic regime equations (19) and (20) can be realized for arbitrary small perturbation $\varepsilon>0, K_{0}>0$. However, the smallest values of $\varepsilon$, and corresponding $K_{0}=K^{\prime}$ that allow for an observable effect has to be found numerically. At first, the external fields are tuned to $K_{0}=K^{\prime}=0.45 \times$ $10^{-4}>0$. In accord with the analytical results stochastic


Figure 3. The same quantities as in figure 2: however, $J=0.2$, $2 \beta=0.1, H_{0 z}=0.2, \varepsilon=0.04$ and $\Omega=100 . \frac{\tau_{0}}{T}=0.1$, $s_{1 z}(0)=0.9, \varphi_{1}(0)=0, s_{2 z}(0)=-0.9, \varphi_{2}(0)=0, T=\frac{2 \pi}{\Omega}$ and $K_{0}=0.32 \times 10^{-4}<K^{\prime}$.


Figure 4. The same quantities as in figure 3: however, $J=0, \beta=0$, $H_{0 z}=0.2, \varepsilon=0.04$ and $\Omega=100 . \frac{\tau_{0}}{T}=0.1, s_{1 z}(0)=0.9$, $\varphi_{1}(0)=0, s_{2 z}(0)=-0.9, \varphi_{2}(0)=0, T=\frac{2 \pi}{\Omega}$ and $K_{0}=0$.
switching of the initial spins' direction occur accompanied by a subsequent long-time stabilization (see figure 2). If the fields are such that $K_{0}<K^{\prime}$ (i.e. $K_{0}$ is very small) switching does not happen (see figure 3), i.e. $s_{1,2 z}$ is still an adiabatic invariant; external fields lead to small fluctuations around the equilibrium state. We note that in figure 3 the anisotropy field is finite but its effect is hardly observable becomes of its smallness $(\beta / J)^{2}=$ 0.06 . The regular (but non-integrable) regime is reached by applying very strong fields $\left(\varepsilon T \gg J, H_{0 z} \gg J s^{z}>2 \beta s^{z}\right)$ (cf equation (3)). In this case no stochastic switching occurs (cf figure 4). The eigenfrequency of the system is given by the constant magnetic field $\omega_{j}\left(S_{i z}\right)=J S_{i z}+H_{0 z}+2 \beta s_{j z} \approx H_{0 z}$. Physically, effects related to the exchange interaction and to


Figure 5. We consider a chain of ten interacting spins. We show here the dynamics of the $z$ components $s_{5, z}$ and $s_{6, z}$ of the spins labelled 5 and 6. Other parameters are $J=0.2,2 \beta=0,3, H_{0 z}=0.2$,
$\varepsilon=0.04$ and $\Omega=100 \cdot \frac{\tau_{0}}{T}=0.1, s_{2 n+1}^{z}(0)=0.9, s_{2 n}^{z}(0)=0.9$, $\varphi_{i}(0)=0, T=\frac{2 \pi}{\Omega}, i=\overline{1, N}$ and $N=10$.
the anisotropy field become negligible and we end up with the familiar resonant switching scheme (this is true only during the time when the external fields are on. Effects of exchange and anisotropy govern the subsequent field-free dynamics. A scheme for a field-induced deflection and freezing has been proposed in [18]).

### 2.4. Finite spin chain

The EOM for a finite spin chain governing the dynamics of each particular spin follows from (2) as

$$
\begin{align*}
& \frac{\mathrm{d} s_{i z}}{\mathrm{~d} t}=-\varepsilon \frac{\partial V_{i}\left(\varphi_{i}, s_{i z}\right)}{\partial \varphi_{i}}, \\
& \frac{\mathrm{~d} \varphi_{i}}{\mathrm{~d} t}=\omega_{i}\left(s_{i-1 z}, s_{i z}, s_{i+1 z}\right)+\varepsilon \frac{\partial V_{i}\left(\varphi_{i}, s_{i z}\right)}{\partial s_{i z}},  \tag{21}\\
& \omega_{i}\left(s_{i-1 z}, s_{i z}, s_{i+1 z}\right)=J s_{i-1 z}+J s_{i+1 z}+2 \beta s_{i z}+H_{0 z}, \\
& \quad i=1, \ldots, N \quad N+1=N
\end{align*}
$$

If the variable field has a spatial extent such that only two spins in the chain, labelled $(k, k+1)$, are affected then we find
$\frac{\mathrm{d} s_{i z}}{\mathrm{~d} t}=-\left(\delta_{i, k}+\delta_{i, k+1}\right) \varepsilon \frac{\partial V_{i}\left(\varphi_{i}, s_{i z}\right)}{\partial \varphi_{i}}$,
$\frac{\mathrm{d} \varphi_{i}}{\mathrm{~d} t}=\omega_{i}\left(s_{i-1 z}, s_{i z}, s_{i+1 z}\right)+\left(\delta_{i, k}+\delta_{i, k+1}\right) \varepsilon \frac{\partial V_{i}\left(\varphi_{i}, s_{i z}\right)}{\partial s_{i z}}$,
$s_{i z}(t)=$ constant $\quad$ if $k \neq i \neq k+1$.
These equations show that the $z$ component of the spins subjected to the pulse have to be determined self-consistently. The dynamics of the oscillation frequency of the spin's transverse components $\dot{\varphi}_{i}, i=1, \ldots, N$ is determined by the effective magnetic field as

$$
\begin{equation*}
\dot{\varphi}_{i}(t)=\omega_{i}^{\mathrm{eff}}=\gamma_{s} H_{i}^{\mathrm{eff}}(t) \tag{23}
\end{equation*}
$$



Figure 6. The time dependence of the $z$ component of the fifth spin for the parameters $J=0.2, H_{0 z}=0.2, \beta=0, H_{0 x}=1$, $s_{2 n+1}^{z}(0)=0.9, s_{2 n}^{z}(0)=-0.9, \varphi_{i}(0)=0, i=\overline{1, N}$ and $N=10$.

Here

$$
\begin{aligned}
& H^{\mathrm{eff}}(t)=\frac{1}{\gamma_{s}}\left[H_{0}+J\left(S_{i-1, z}+S_{i+1, z}\right)\right. \\
& \left.\quad+\left(2 \beta-\left(\delta_{i, k}+\delta_{i, k+1}\right) \frac{H_{0 x}(t)}{S_{i \perp}} \cos \left(\varphi_{i}\right)\right) S_{i, z}\right]
\end{aligned}
$$

This indicates that the spins subject to the pulses exchange energy with their nearest neighbours (whose $z$ components are nevertheless constant). This process depends on the values of the $z$ components and on the effective frequency $\omega_{i}^{\text {eff }}(t) s_{i}^{z}(t)$; a demonstration of this phenomena is shown in figure 5.

A further tool for controlling the diffusion process is to apply a constant field along the $x$ axis. The system dynamics is then chaotic, even without the periodic series of pulses [4,5]. Therefore, the $z$ component of the spin is not an adiabatic invariant and the mechanism of dynamical freezing (DF) discussed above does not work. Figure 6 illustrates that, if the amplitude of the magnetic field applied along the $x$ axis is strong enough $H_{0 x}>H_{0 z}$, then the longitudinal component of the spin performs fast oscillations. In the other opposite case $H_{0 x}<H_{0 z}$ the orientation of the spin can be deflected but DF again is not possible (cf figure 7).

After deflection of the spin to a desired angle, one can completely freeze its orientation. The key point is the fact that, in the absence of pulses, the $z$ component of the spin projection is an integral of motion. The system of equations in this case has the form

$$
\begin{gather*}
\frac{\mathrm{d} s_{j x}}{\mathrm{~d} t}=-J\left(s_{j-1, z}+s_{j+1, z}\right) s_{j y}-H_{0 z} s_{j y} \\
\frac{\mathrm{~d} s_{j y}}{\mathrm{~d} t}=J\left(s_{j-1, z}+s_{j+1, z}\right) s_{j x}+H_{0 z} s_{j x}  \tag{24}\\
\frac{\mathrm{~d} s_{j z}}{\mathrm{~d} t}=0
\end{gather*}
$$



Figure 7. The same as in figure 6: however, the parameters are changed to $J=0.2, H_{0 z}=0.2, H_{0 x}=0.01, s_{2 n+1}^{z}(0)=0.9$, $s_{2 n}^{z}(0)=-0.9, \varphi_{i}(0)=0, i=\overline{1, N}$ and $N=10$.

For simplicity we assumed here that $\beta=0$. After solving (24) one obtains

$$
\begin{gather*}
s_{j x}=s_{j x}\left(t_{0}\right) \cos \left(\omega_{j 0} t+\varphi_{0}\right) \\
s_{j y}=s_{j y}\left(t_{0}\right) \sin \left(\omega_{j 0} t+\varphi_{0}\right), \quad s_{j z}=s_{j z}\left(t_{0}\right), \tag{25}
\end{gather*}
$$

where $\omega_{j}\left(s_{j-1 z}(0) ; s_{j+1 z}(0)\right)=J\left(s_{j-1 z}(0)+s_{j+1 z}(0)\right)+$ $H_{0 z}=\omega_{j 0}$, and $s_{j z}\left(t_{0}\right)$ corresponds to the desired orientation of the spin, achieved after the action of the pulses in the time interval $0<t<t_{0}$ (cf figure 8 ).

### 2.5. Arnold diffusion

Results of section 2.4 show that, even in the case of a long spin chain, the orientation of spins can still be controlled. This follows from resonance overlapping and the existence of diffusion. However, the question of what kind of diffusion we have is still outstanding. If the dimension of the system is more than $N>2$, the dynamics is much more involved and the emergence of new physical phenomena is expected. We recall the key idea of KAM theory: the size of the destroyed torus is small and the domain of their location is surrounded by an invariant torus. This situation changes if an invariant torus crosses the domain of the destroyed torus location. This is possible if and only if $N>2$. The phenomenon of universal diffusion along the net formed due to the torus crossing was discovered by Arnold [6]. Here we consider the mechanism of the formation of the stochastic net in the case of a spin chain:

$$
\begin{equation*}
H=H_{0}\left(s_{1}^{z}, \ldots, s_{N}^{z}\right)+\varepsilon V\left(\varphi_{1}, \ldots, \varphi_{N}\right) \tag{26}
\end{equation*}
$$

Note, the frequencies of the unperturbed motion on the $N$ dimensional torus is a function of the three actions

$$
\begin{align*}
& \omega_{i}\left(s_{i-1}^{z}, s_{i}^{z}, s_{i+1}^{z}\right)=J s_{i-1}^{z}+J s_{i+1}^{z}+2 \beta s_{i}^{z}+H_{0 z} \\
& \quad i=\overline{1, N} \tag{27}
\end{align*}
$$



Figure 8. The $z$ components of the spins labelled 5 and 6 for the parameters: $J=0.2, H_{0 z}=0.2, H_{0 x}=0.01, s_{2 n+1}^{z}(0)=0.9$, $s_{2 n}^{z}(0)=-0.9, \varphi_{i}(0)=0, i=\overline{1, N}$ and $N=10$.

We collect the resonant torus defined by the condition:

$$
\begin{equation*}
\sum_{j=1}^{N} n_{j} \omega_{j}=0 \tag{28}
\end{equation*}
$$

where $n_{j}$ are integer numbers. For each set of numbers there exists a multitude of solutions $s_{z}^{0} \equiv\left(s_{1}^{z(0)}, \ldots, s_{N}^{z(0)}\right)$. Each solution determines the resonant torus. For the formation of the Arnold diffusion the absences of degeneracy is essential:

$$
\begin{equation*}
\operatorname{det}\left|\frac{\partial^{2} H_{0}}{\partial s_{i}^{z} \partial s_{j}^{z}}\right| \neq 0, \quad i, j=\overline{1, N} \tag{29}
\end{equation*}
$$

In the case of our system due to the form of the matrix

$$
\frac{\partial^{2} H_{0}}{\partial s_{i}^{z} \partial s_{j}^{z}}=\left(\begin{array}{ccccc}
2 \beta & J & 0 & 0 & \cdot  \tag{30}\\
J & 2 \beta & J & 0 & \cdot \\
0 & J & 2 \beta & J & \cdot \\
0 & 0 & J & 2 \beta & \cdot \\
\cdot & \cdot & \cdot & \cdot & \cdot
\end{array}\right)
$$

the condition of the absence of degeneration (29) leads to the polynomial expression:

$$
\begin{equation*}
\operatorname{det}\left|\frac{\partial^{2} H_{0}}{\partial s_{i}^{z} \partial s_{j}^{2}}\right|=G(J, \beta, N) \neq 0 \tag{31}
\end{equation*}
$$

The explicit form of the expression (31) also depends on the system's size (in addition to the dependence on the parameters $\beta, J)$. For large systems we obtain the following asymptotic expressions:

$$
G(J, \beta, N)= \begin{cases}J^{N} & \text { if } J>\beta  \tag{32}\\ 2^{N} \beta^{N} & \text { if } J<\beta\end{cases}
$$

From this relation we can conclude that the universal diffusion is possible for any nonzero $J, \beta$, and identify the numerical
results obtained for the spin chain with the Arnold diffusion. For an analytical estimation we consider the minimal possible dimension. Therefore, in what follows, without loss of generality we shall restrict ourselves to the case $N=3$. From equation (28) we find

$$
\begin{equation*}
n_{1} \omega_{1}+n_{2} \omega_{2}+n_{3} \omega_{3}=0, \tag{33}
\end{equation*}
$$

where each frequency depends on the three actions according to (27). In the frequency space $\left(\omega_{1}, \omega_{2}, \omega_{3}\right)$ equation (33) determines a family of surfaces. On the energy surface we have

$$
\begin{equation*}
H_{0}\left(s_{1}^{z}, s_{2}^{z}, s_{3}^{z}\right)=E . \tag{34}
\end{equation*}
$$

This equation is also the equation determining the surface (33). Therefore, the resonant torus have a common part along the curves, defined as the solutions of the set of equations (33) and (34). The time-dependent perturbation leads to a widening of these curves and to the formation of the stochastic net.

In order to provide a topological interpretation of this phenomenon we will consider the simplest case of three spins. In this case the explicit form of equations (33) and (34) is

$$
\begin{equation*}
(J+2 \beta)\left(\omega_{1}^{2}+\omega_{2}^{2}+\omega_{3}^{2}\right)-2 \omega_{1} \omega_{2}-2 \omega_{1} \omega_{3}-2 \omega_{2} \omega_{3}=E \tag{35}
\end{equation*}
$$

and

$$
\begin{equation*}
n_{1} \omega_{1}+n_{2} \omega_{2}+n_{3} \omega_{3}=0 \tag{36}
\end{equation*}
$$

Here $E=\left(8 \beta^{2}+4 \beta J-4 J^{2}\right) H_{0}-3 H_{0 z}^{2}(2 \beta-J)$ is a rescaled energy. From equation (36) one can exclude frequency $\omega_{1}=-\frac{n_{2}}{n_{1}} \omega_{2}-\frac{n_{3}}{n_{1}} \omega_{3}$ and rewrite equation (35) as a function of the two frequencies $\left(\omega_{3}, \omega_{2}\right)$. Clearly, the shape of the implicit plot for $\omega_{3}\left(\omega_{2}\right)$ depends on the values of parameters $\left(n_{1}, n_{2}, n_{3}\right)$. Therefore, we expect that the $\omega_{3}\left(\omega_{2}\right)$, plotted for different inner resonances $\left(n_{1}, n_{2}, n_{3}\right)$, should cross at some points. Now one can construct implicit plots expressing frequencies as a function of each other for different resonances (cf figure 9). We see that, at some points, trajectories cross each other. Due to topological reasons, such nodal points are possible if and only if the system dimension is at least $N=3$ or higher. Nodal points are crossing points between different resonances. If an external adiabatic perturbation is applied the dynamic near the nodal points becomes unpredictable and this leads to the Arnold diffusion [6].

In the general dimensional case $(N \gg 1)$, the geometrical interpretation is less illustrative and much more complicated. Since we have to deal with $(N-1)$-dimensional hypercurves in the $N$-dimensional hyperspace, the basic concept is, however, the same [20]. This conclusion manifests fundamental features of the multidimensional nonlinear dynamical systems. The diffusive motion of the system in the stochastic net is named as Arnold diffusion. Therefore, the diffusion equation (16) is still justified. However, the coefficient of diffusion for Arnold diffusion is defined by an expression other than equation (16), namely [6]

$$
\begin{equation*}
D_{\mathrm{A}}=E^{2} \varepsilon H_{0 z} \mathrm{e}^{-1 / \varepsilon^{a(N)}} . \tag{37}
\end{equation*}
$$

Here $E$ is the system's energy, $a(N)$ is a dimensionaldependent scaling constant with an upper limit determined by the Arnold inequality relation [6]:

$$
\begin{equation*}
a(N)<\frac{2}{6 N(N-1)+3 N+14} \tag{38}
\end{equation*}
$$



Figure 9. Topological structure of the inner resonances on the frequency plane $\left(\omega_{3}, \omega_{2}\right)$ plotted for different resonances: $\left(n_{2}=n_{1}, n_{3}=n_{1}\right),\left(n_{2}=2 n_{1}, n_{3}=3 n_{1}\right),\left(n_{2}=3 n_{1}, n_{3}=\right.$ $\left.n_{1}\right),\left(n_{2}=5 n_{1}, n_{3}=7 n_{1}\right)$. Further values: $J=0.2,2 \beta=0.3$.

Obviously, for $N \gg 1, a(N) \mapsto 0$ and the coefficient of the Arnold diffusion takes the simpler form

$$
\begin{equation*}
D_{\mathrm{A}}=\frac{1}{e} \varepsilon E^{2} H_{0 z} \tag{39}
\end{equation*}
$$

Comparing equation (39) with the diffusion coefficient obtained for the case of two spins, i.e. equation (16), we find

$$
\begin{equation*}
\frac{D_{\mathrm{A}}}{D}=\frac{4 E^{2} H_{0 z}}{e \varepsilon T} . \tag{40}
\end{equation*}
$$

This relation is important in that it delivers information on when the mechanism of stochastic switching and dynamical freezing are more efficient $\frac{D_{A}}{D}>1$ for a long spin chain, as compared to the case of a pair of spins.

### 2.6. Role of anisotropy field

Here we discuss the connection between the anisotropy field and Arnold diffusion. For the Arnold diffusion to occur the Jacobi matrix has to be non-degenerate. We note, however, that the Jacobi matrix becomes degenerate in some cases if the anisotropy field is absent, as can be inferred from the structure
of the Jacobi matrix. For example, in the simplest case of three spins
$\operatorname{det}\left|\frac{\partial^{2} H_{0}}{\partial S_{i}^{z} \partial S_{j}^{z}}\right|=-4 J^{2} \beta+8 \beta^{3} \neq 0, \quad$ if $\beta \neq 0$.
Evaluating the determinant for different numbers of spins we find that with the anisotropy field being applied it is always non-degenerate, while for a particular $N$, it becomes degenerate in the absence of the anisotropy field.

## 3. Low temperature limit

In this section we consider the dynamics in the continuous limit. By a proper choice of pulse parameters we were able to deflect the spin orientation diffusively to a desired angle. Switching off the pulses, the dynamics remain quasi-frozen (is equivalent to the spin $z$ component being an integral of motion). The question we pose here is that, what happens if upon stochastic switching and freezing we apply a constant magnetic field along the $x$ axis. We recall that applying a constant field to the equilibrium (initial) state along the $x$ axis invalidates the use of the KAM theory and the mechanism of SC does not work (cf equation (5)). Before we deal with this problem in more detail we set the limits of continuous approximation. Due to the constant magnetic field, applied along the $x$ axis, the $s_{z}$ component is not an integral of motion any more. Therefore, excitations similar to spin waves propagate along the spin chain. These waves are not completely analogous to spin waves because the nonconservation of $s_{z}(t)$ is not related to flip-flop processes, but requires a transversal magnetic field. However, the wavelength of such excitations can be evaluated in a manner similar to the spin waves' case [24]. If the wavelength is larger than the distance between the spins $\lambda \gg a$ a continuous treatment is justified. Taking into account the expression for the wave frequency

$$
\begin{equation*}
\omega=\frac{4|J| S}{\hbar} \frac{2 \pi}{\lambda} a, \tag{41}
\end{equation*}
$$

One concludes that the validity of the continuous approximation depends on the temperature

$$
\begin{equation*}
T \ll \frac{J \hbar}{k_{\mathrm{B}}} . \tag{42}
\end{equation*}
$$

Here $k_{\mathrm{B}}$ is the Boltzmann constant. Thus, the continuous approximation corresponds to a low temperature approximation. For anti-ferromagnetic materials $\mathrm{FeCl}_{2}$ or $\mathrm{CoCl}_{2}$ we have $J=1.23 \times 10^{12} \mathrm{~Hz}$, from (42) we infer for a temperature regime of the continuous model $T<3 \mathrm{~K}$.

Returning back to the spin chain in a static magnetic field along the $x$ axis, the EOM is

$$
\begin{gathered}
\frac{\mathrm{d} s_{j x}}{\mathrm{~d} t}=-J\left(s_{j-1, z}+s_{j+1, z}\right) s_{j y}-H_{0 z} s_{j y} \\
\frac{\mathrm{~d} s_{j y}}{\mathrm{~d} t}=J\left(s_{j-1, z}+s_{j+1, z}\right) s_{j x}+H_{0 z} s_{j x}-H_{0 x} s_{j z} \\
\frac{\mathrm{~d} s_{j z}}{\mathrm{~d} t}=H_{0 x} s_{j y}
\end{gathered}
$$

Considering that

$$
\begin{gather*}
s_{j x} \rightarrow s_{x}(x ; t), \quad s_{j y} \rightarrow s_{y}(x ; t),  \tag{44}\\
s_{j z} \rightarrow s_{z}(x ; t) \\
s_{j-1 ; z}=s_{x}(x, t)-a \frac{\partial s_{z}(x, t)}{\partial x}+\frac{a^{2}}{2} \frac{\partial^{2} s_{x, t}}{\partial t^{2}}, \\
s_{j+1 ; z}=s_{x}(x, t)+a \frac{\partial s_{z}(x, t)}{\partial x}+\frac{a^{2}}{2} \frac{\partial^{2} s_{x, t}}{\partial t^{2}}, \tag{45}
\end{gather*}
$$

from (43) we deduce that

$$
\begin{gather*}
\frac{\partial s_{x}}{\partial t}=-J s_{y}\left(2 s_{z}+\frac{\partial^{2} s_{z}}{\partial x^{2}} a^{2}\right)-H_{0 z} s_{y} \\
\frac{\partial s_{x}}{\partial t}=J s_{x}\left(2 s_{z}+\frac{\partial^{2} s_{z}}{\partial x^{2}} a^{2}\right)-H_{0 x} s_{z}+H_{0 z} s_{x}  \tag{46}\\
\frac{\partial s_{z}}{\partial t}=H_{0 x} s_{y}
\end{gather*}
$$

In the low temperature regime we can neglect quadratic terms in (46) and obtain

$$
\begin{gather*}
\dot{x}=-\delta y-\gamma y z, \quad \dot{y}=\delta x-z+\gamma x z, \\
\dot{z}=y . \tag{47}
\end{gather*}
$$

Here we introduced the following notations:

$$
\begin{aligned}
\delta=\frac{H_{0 z}}{H_{0 x}}, & \gamma=\frac{2 J}{H_{0 x}}, \\
s_{x}=x, & t \rightarrow H_{0 x} t \\
s_{y}=y, & s_{z}=z
\end{aligned}
$$

Equation (47) is derived in the absence of an anisotropy field. However, for the role of the anisotropy field we remark the following: the Zeeman field applied along the $z$ axis is very strong $H_{0}^{z}>J\left|S_{i-1}^{z}\right|+J\left|S_{i+1}^{z}\right|+2 \beta\left|S_{i}^{z}\right|$. The eigenfrequency is constant and we have no effect of a dynamical shift $\omega_{i}\left(S_{i-1}^{z}, S_{i+1}^{z}, S_{i}^{z}\right)=J S_{i-1}^{z}+J S_{i+1}^{z}+2 \beta S_{i}^{z}+H_{0}^{z} \approx H_{0}^{z}$. Inclusion of a finite anisotropy field leads to a rescaling of the small parameter $\gamma$ in equation (47), i.e. $\gamma=\frac{2 J}{H_{0 x}} \rightarrow \frac{2 J+2 \beta}{H_{0 x}}$. Hence, we conclude that in this particular case the anisotropy field has no principal dynamical effect. Equation (47) with rescaled parameter $\gamma=\frac{2 J+2 \beta}{H_{0 x}}$ is still valid in the presence of an anisotropy field.

When solving (47), we assume for the initial values the spin orientations achieved after the action of pulses. In order to obtain analytical solutions we utilize the canonical perturbation theory (cf, e.g., [25]). The parameter $\gamma$ is assumed to be small. The first step is to rewrite (47) in a canonical form. This can be done using the following transformation (for details, see the appendix):

$$
\begin{gather*}
x_{1}=2 \delta x-2 z, \quad y_{1}=2 \lambda y, \quad z_{1}=\frac{1}{\delta} x+z,  \tag{48}\\
\lambda=\sqrt{1+\delta^{2}} .
\end{gather*}
$$



Figure 10. The longitudinal $s_{z}$, and the transversal $s_{\perp}=\sqrt{s_{x}^{2}+s_{y}^{2}}$ spin components as a function of time. $J=0.05, H_{0 z}=1$ and $H_{0 x}=0.1$. Graph (a) shows the analytical solutions given by equation (52), whereas in graph (b) the numerical integration of the system of equation (47) is depicted.

Equations (47) assume then the form

$$
\begin{gather*}
\dot{x_{1}}=-\lambda y_{1}+\frac{\delta \gamma}{2 \lambda^{3}} y_{1} x_{1}-\frac{\gamma \delta^{3}}{\lambda^{3}} y_{1} z_{1} \\
\dot{y_{1}}=\lambda x_{1}+\frac{\gamma \delta\left(\delta^{2}-1\right)}{\lambda^{3}} x_{1} z_{1}-\frac{\gamma \delta}{2 \lambda^{3}} x_{1}^{2}+\frac{2 \gamma \delta^{3}}{\lambda^{3}} z_{1}^{2}  \tag{49}\\
\dot{z_{1}}=\frac{\gamma}{4 \lambda^{3} \delta} y_{1} x_{1}-\frac{\gamma \delta}{2 \lambda^{3}} y_{1} z_{1}
\end{gather*}
$$

We seek a solution of (49) having the structure

$$
\begin{align*}
& x_{1}=C x_{1}^{1}+C^{2} x_{1}^{(2)}+C^{3} x_{1}^{(3)}+C^{4} x_{1}^{(4)} \cdots \\
& y_{1}=C y_{1}^{1}+C^{2} y_{1}^{(2)}+C^{3} y_{1}^{(3)}+C^{4} y_{1}^{(4)} \cdots  \tag{50}\\
& z_{1}=C z_{1}^{1}+C^{2} z_{1}^{(2)}+C^{3} z_{1}^{(3)}+C^{4} z_{1}^{(4)} \cdots,
\end{align*}
$$

and in addition we use the re-scaled time

$$
\begin{equation*}
t=\frac{\tau}{\lambda}\left(1+h_{2} C^{2}+h_{3} C^{3}+\cdots\right) \tag{51}
\end{equation*}
$$

With an accuracy up to third order in $\gamma$ we find the solution of (47) to be

$$
\begin{aligned}
s_{x}= & \frac{\delta \cos \tau}{\lambda}+\frac{\gamma\left(1+2 \delta^{2}\right) \sin ^{2}(\tau)}{2 \lambda^{4}} \\
& -\frac{\gamma^{2} \delta\left(9+4 \delta^{2}\right) \cos (\tau) \sin ^{2}(\tau)}{16 \lambda^{7}} \\
s_{y}= & \sin (\tau)-\frac{\gamma \delta \cos (\tau) \sin (\tau)}{\lambda^{3}} \\
& +\frac{\gamma^{2}\left(1+4 \delta^{2}\right)(-5 \sin (\tau)+3 \sin (3 \tau))}{64 \lambda^{6}} \\
s_{z}= & -\frac{\cos \tau}{\lambda}-\frac{\gamma \delta \sin ^{2}(\tau)}{2 \lambda^{4}} \\
& -\frac{\gamma^{2}\left(-1+4 \delta^{2}\right) \cos (\tau) \sin ^{2}(\tau)}{16 \lambda^{7}} .
\end{aligned}
$$

Figure 10 demonstrates that these analytical solutions are in good agreement with the exact numerical simulation of the system (47). Figure 10 shows that a constant magnetic field results in oscillations of the spin's longitudinal component in a controlled manner. If the amplitude of the magnetic field is small, nonlinear effects become more important (cf figure 11).

## 4. Quantum mechanical consideration and the problem of DF

As stated above, the classical analysis is useful if the atoms in the chain have a large magnetic moment. In fact, for a finite chain of manganese (Mn) atoms [1] the classical approach proved to be adequate [2]. This situation changes, however, for small spins where the dynamics becomes dominated by quantum effects. What is needed for our quantum consideration is the structure of the energy spectrum in the regime where the underlying classical dynamics is chaotic [4]. The point of interest here is that whether quantum effects invalidate the SC and, in particular, die on dynamical freezing. To this end we use the concept of quantum geometry [26, 28] in the way done in $[8,9]$ to study DF. Let us consider two quantum states $\Psi_{1}$ and $\mathrm{e}^{\mathrm{i} \varphi} \Psi_{1}$. The distance in Hilbert space between them can be characterized by the quantity [26-28]

$$
D_{1}\left(\Psi_{1}, \Psi_{2}\right)=\min _{\varphi}\left\|\Psi_{1}-\mathrm{e}^{\mathrm{i} \varphi} \Psi_{2}\right\|
$$

The minimal phase $\varphi_{m}$ is found by exterminating $\left\|\Psi_{1}-\mathrm{e}^{\mathrm{i} \varphi} \Psi_{2}\right\|$ and noting that $\left\|\Psi_{1}-\Psi_{2}\right\|=\left\langle\Psi_{1} \mid \Psi_{2}\right\rangle^{1 / 2}$, which yields

$$
\begin{equation*}
\exp \left(\mathrm{i} \varphi_{m}\right)=\frac{\left\langle\Psi_{1} \mid \Psi_{2}\right\rangle}{\left|\left\langle\Psi_{1} \mid \Psi_{2}\right\rangle\right|} \tag{53}
\end{equation*}
$$

Therefore, we write for the distance $D_{1}$

$$
D_{1}\left(\Psi_{1}, \Psi_{2}\right)=\sqrt{2-2\left|\left\langle\Psi_{1} \mid \Psi_{2}\right\rangle\right|}
$$



Figure 11. The same as in figure 10 with the same meaning of the labels. The parameters of the system are, however, changed to $J=0.05$, $H_{0 z}=0.2$ and $H_{0 x}=0.1$.

For the same purpose as for $D_{1}$, one may also use the FubiniStudy metric [27]:

$$
\begin{equation*}
D_{2}^{2}(\Psi(t), \Psi(0))=4\left(1-|\langle\Psi(t) \mid \Psi(0)\rangle|^{2}\right) . \tag{54}
\end{equation*}
$$

In both approaches the key quantity is the so-called Bargmann angle [8]:

$$
\begin{equation*}
\cos \theta_{\mathrm{B}}(t)=A(t)=|\langle\Psi(0) \mid \Psi(t)\rangle| . \tag{55}
\end{equation*}
$$

In analogy to the classical deflection in quantum control problems, the Bargmann angle can be considered as the 'quantum deflection'. Essential for further progress is our (classical) finding that SC and DF occur in the classical chaotic regime. This calls for the use of random matrix theory (RMT) to inspect the quantum dynamics [29-31]. To this end we write (2) as

$$
\begin{equation*}
\hat{H}(t)=\hat{H}_{0}+\hat{V}(t), \tag{56}
\end{equation*}
$$

where $\hat{H}_{0}$ is time-independent. Now we employ the established Floquet-operator method [30] and the quantum map and infer for the Bargmann angle

$$
\begin{equation*}
A^{2}(t)=\frac{1}{N^{2}}\left(N+\sum_{\substack{n, m=1 \\ n \neq m}}^{N} \cos \left[t\left(\varphi_{n}-\varphi_{m}\right)\right]\right) \tag{57}
\end{equation*}
$$

Here $\varphi_{n}$ stands for the eigenphases of the Floquet operators and $N$ is the Hilbert space dimension. From this relation it is evident that, starting at $t=0$ with two completely coherent states, i.e. $A(t=0)=1$, decoherence sets in for $t \neq 0$. To put the classical predictions of section 3 into a quantum perspective we note the following: In the classical regular regime, as identified above, the time-dependent perturbation $\hat{V}(t)$ acts adiabatically and does not alter the structure or the symmetry of the quantum spectrum. In this case we expect the Bargmann angle to be time-periodic with typical quantum revivals. In contrast, in the classical chaotic regime, $\hat{V}(t)$ changes qualitatively the quantum spectrum.

Starting from the (classically) regular case we conclude that, if pair excitations are neglected, then the energy spectrum of the unperturbed part has the form

$$
\begin{equation*}
E_{n}=(n-1-N / 2) H_{0 z}+\frac{1}{4} J(N-4 n+3)+\frac{\beta N}{4} . \tag{58}
\end{equation*}
$$

With this spectrum we infer for the Bargmann angle (equation (57)) the expression

$$
\begin{align*}
A^{2}(t) & =\frac{1}{N^{2}}\left(\frac{\sin (N+1 / 2) t-\sin (t / 2)}{2 \sin (t / 2)}\right)^{2} \\
+ & \frac{1}{N^{2}}\left(\frac{\cos (t / 2)-\cos (N+1 / 2) t}{2 \sin (t / 2)}\right)^{2} . \tag{59}
\end{align*}
$$

As is evident from figure 12 , if the underlying classical dynamics is regular, the time dependence of the Bargmann angle is periodic and the system is characterized by quantum revivals.

To deal quantum mechanically with the classically chaotic regime we follow [30] and employ a Gaussian orthogonal ensemble [30]:

$$
\begin{equation*}
P\left(\varphi_{1}, \ldots, \varphi_{N}\right)=\prod_{n>m}\left(\varphi_{n}-\varphi_{m}\right) \exp \left(-\sum_{n=1}^{N} \varphi_{n}^{2}\right) . \tag{60}
\end{equation*}
$$

We note here that in general the distribution function (60) includes correlations between all $N$ levels. If the number of correlated levels is $n$, then the $n<N$ level-correlated distribution function is

$$
\begin{align*}
& P_{n}\left(\varphi_{1}, \ldots, \varphi_{n}\right) \\
& \quad=\frac{N!}{(N-n)!} \int P_{n}\left(\varphi_{1}, \ldots, \varphi_{N}\right) \mathrm{d} \varphi_{n+1} \cdots \mathrm{~d} \varphi_{N} . \tag{61}
\end{align*}
$$

The structure of the expression (57) suggests that the secondorder correlated level distribution function $P_{2}\left(\varphi_{n}, \varphi_{m}\right)$ is sufficient (each term in the sum contains two phases). Upon straightforward calculations we reduced $P_{2}\left(\varphi_{n}, \varphi_{m}\right)$ to
$P_{2}\left(\varphi_{n}, \varphi_{m}\right)=K_{N}\left(\varphi_{n}, \varphi_{n}\right) K_{N}\left(\varphi_{m}, \varphi_{m}\right)$
$-K_{N}\left(\varphi_{n}, \varphi_{m}\right) K_{N}\left(\varphi_{m}, \varphi_{n}\right)$,


Figure 12. Bargmann angle $\theta_{\mathrm{B}}(t)$ as a function of re-scaled time $t \equiv t /\left(H_{0 z}+J\right)$ and $H_{0 z}=J=0,2$ calculated from (59).
with

$$
K\left(\varphi_{n}, \varphi_{m}\right)=\sum_{k=1}^{N} \phi_{k}\left(\varphi_{n}\right) \phi_{k}\left(\varphi_{n}\right)
$$

and

$$
\phi_{k}(\varphi)=\frac{1}{\left(2^{n} n!\sqrt{\pi}\right)^{1 / 2}} H_{k}(\varphi) \exp \left[-\frac{\varphi^{2}}{2}\right]
$$

$H_{n}(\varphi)$ are Hermite polynomials. For $\left\langle A^{2}(t)\right\rangle_{P_{2}}$ we find (for $N \gg 1$ )

$$
\begin{align*}
& \left\langle A^{2}(t)\right\rangle_{P_{2}}=C \exp \left[-t^{2} / 2\right]\left\{\sum_{n, m}^{N} L_{n}^{0}\left(t^{2} / 2\right) L_{\mathrm{m}}^{0}\left(t^{2} / 2\right)\right. \\
& -\frac{\sqrt{\pi}}{4} \sum_{m=1}^{N} \sum_{n=1}^{N} \frac{n!}{2^{m-n} m!} t^{m-n}\left(L_{n}^{m-n}\left(t^{2} / 2\right)\right)^{2} \\
& \left.\quad-\frac{\sqrt{\pi}}{4} \sum_{m=1}^{N} \sum_{n=1}^{m-1} \frac{n!}{2^{m-n} m!} t^{m-n-1}\left(L_{n}^{m-n-1}\left(t^{2} / 2\right)\right)^{2}\right\} \tag{62}
\end{align*}
$$

where $L_{n}^{m}\left(t^{2} / 2\right)$ are Laguerre polynomials. The constant $C$ we find from the condition $\left\langle A^{2}(0)\right\rangle_{P_{2}}=1$. Dynamical freezing (DF) means then a stabilization over time of the quantum distance $[8,9]$ quantified by the Bargmann angle. To test for this situation we numerically solve for (62); a typical example is shown in figure 13. These calculations are performed for parameters appropriate for the classically chaotic regime, e.g. those of figure 2. The interpretation of figure 13 is that SC drives the system diffusively to the target state $|\Psi(t)\rangle$, which in this case is orthogonal to $|\Psi(0)\rangle$. The Bargmann angle is therefore deflected within a time $t_{\mathrm{D}}$ determined by the diffusion constant to the value $\theta_{\mathrm{B}}\left(t_{\mathrm{D}}\right) \sim \pi / 2$. DF is then shown by a small variation of $\theta_{\mathrm{B}}\left(t_{\mathrm{D}}\right)$ for $t>t_{\mathrm{D}}$.

### 4.1. Conclusions

For an exchange-coupled, nonlinear spin chain and in the presence of a (uniaxial) magnetic anisotropy and external driving fields, stochastic switching is possible if the field parameters are chosen such that the underlying classical dynamics is chaotic. The switching mechanism is identified to be Arnold-type diffusion. This we concluded analytically and substantiated by full numerical semiclassical and quantum calculations. We also inspected the possibility of dynamical


Figure 13. The time evolution of the Bargmann angle $\theta_{\mathrm{B}}(t)$, as calculated using equation (62) and averaged over the random quantum phases. The parameters of the driving fields and the spin chains are the same as in figure 2, i.e. we are in the stochastic switching regime.
freezing, i.e. stabilizing the target state beyond the switching time.

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## Appendix

The canonical Lyapunov system has the general structure

$$
\begin{gather*}
\dot{x}=-\lambda y+X\left(x, y, z_{1}, \ldots, z_{m}\right) \\
\dot{y}=\lambda x+Y\left(x, y, z_{1}, \ldots, z_{m}\right) \\
\dot{z}_{s}=\sum_{j=1}^{m} b_{s j}+Z_{s}\left(x, y, z_{1}, \ldots, z_{m}\right)  \tag{A.1}\\
(s=1,2, \ldots, m ; m=n-2)
\end{gather*}
$$

We seek a reduction of this system of equations using the canonical transformation

$$
\begin{gather*}
\dot{x}=-\delta y-\gamma y z, \quad \dot{y}=\delta x-z+\gamma x z  \tag{A.2}\\
\dot{z}=y
\end{gather*}
$$

We are interested in the linear part of equation (A.2), i.e.

$$
\begin{equation*}
\dot{x}=-\delta y, \quad \dot{y}=\delta x-z, \quad \dot{z}=y \tag{A.3}
\end{equation*}
$$

From the coefficients of equation (A.3) we can construct the following matrix:

$$
a=\left(\begin{array}{ccc}
0 & -\delta & 0  \tag{A.4}\\
\delta & 0 & -1 \\
0 & 1 & 0
\end{array}\right)
$$

We consider now the linear transformation

$$
\begin{align*}
& \xi_{1}=\gamma_{11} x+\gamma_{12} y+\gamma_{13} z, \\
& \xi_{2}=\gamma_{21} x+\gamma_{22} y+\gamma_{23} z,  \tag{A.5}\\
& \xi_{3}=\gamma_{31} x+\gamma_{32} y+\gamma_{33} z .
\end{align*}
$$

In the new variables equation (A.3) is cast as

$$
\begin{equation*}
\frac{\mathrm{d} \xi_{i}}{\mathrm{~d} t}=\lambda_{i} \xi_{i} \quad i=1,2,3 \tag{A.6}
\end{equation*}
$$

Taking equations (A.5) and (A.3) into account we infer from equation (A.6) that

$$
\begin{align*}
& \left(a_{11}-\lambda_{i}\right) \gamma_{i 1}+a_{21} \gamma_{i 2}+a_{31} \gamma_{i 2}=0, \\
& a_{12} \gamma_{i 1}+\left(a_{22}-\lambda_{i}\right) \gamma_{i 2}+a_{32} \gamma_{i 3}=0,  \tag{A.7}\\
& a_{13} \gamma_{i 1}+a_{23} \gamma_{i 2}+\left(a_{33}-\lambda_{i}\right) \gamma_{i 3}=0 .
\end{align*}
$$

Equating the determinant to zero

$$
\left|\begin{array}{ccc}
-\lambda_{i} & -\delta & 0  \tag{A.8}\\
\delta & -\lambda_{i} & -1 \\
0 & 1 & -\lambda_{i}
\end{array}\right|=0,
$$

we find

$$
\begin{equation*}
\lambda_{1,2}= \pm \mathrm{i} \lambda, \quad \lambda_{3}=0, \quad \lambda=\sqrt{1+\delta^{2}} . \tag{A.9}
\end{equation*}
$$

According to (A.7) this gives the following solutions for the matrix:

$$
\begin{array}{lll}
\gamma_{11}=-\delta, & -\gamma_{12}=-\mathrm{i} \lambda, & \gamma_{13}=1 ; \\
\gamma_{21}=-\delta, & -\gamma_{22}=-\mathrm{i} \lambda, & \gamma_{23}=1 ;  \tag{A.10}\\
\gamma_{31}=-\frac{1}{\delta}, & -\gamma_{22}=0, & \gamma_{33}=1 .
\end{array}
$$

So the canonical transformation (A.5) has the form

$$
\begin{gather*}
\xi_{1}=-\delta x-\mathrm{i} \lambda y+z ; \quad \xi_{2}=-\delta x+\mathrm{i} \lambda y+z \\
\xi_{3}=-\frac{1}{\delta} x+z \tag{A.11}
\end{gather*}
$$

The inverse transformation is

$$
\begin{gather*}
x=\frac{\delta}{\lambda^{2}}\left(\xi_{3}-\frac{\xi_{1}+\xi_{2}}{2}\right), \quad y=\frac{\mathrm{i}}{2 \lambda}\left(\xi_{1}+\xi_{2}\right),  \tag{A.12}\\
z=\frac{\xi_{1}+\xi_{2}+2 \delta^{2} \xi_{3}}{2 \lambda^{2}} .
\end{gather*}
$$

Using equation (A.10) we obtain then for equations of motion in the variable $\xi_{i}$ :

$$
\begin{equation*}
\dot{\xi}_{1}=\mathrm{i} \lambda \xi_{1}, \quad \dot{\xi}_{2}=-\mathrm{i} \lambda \xi_{2}, \quad \dot{\xi}_{3}=0 \tag{A.13}
\end{equation*}
$$

To reduce our system of equations to the canonical form, one more transformation is needed:

$$
\begin{equation*}
x_{1}=-\left(\xi_{1}+\xi_{2}\right), \quad y_{1}=i\left(\xi_{1}-\xi_{2}\right), \quad z_{1}=\xi_{3} \tag{A.14}
\end{equation*}
$$

Taking equations (A.10) and (A.11) into account we obtain

$$
\begin{equation*}
x_{1}=2 \delta x-2 z ; \quad y_{1}=2 \lambda y ; \quad z_{1}=\frac{1}{\delta} x+z \tag{A.15}
\end{equation*}
$$

The inverse transformation is

$$
\begin{gather*}
x=\frac{\delta}{\lambda^{2}}\left(z_{1}+\frac{x_{1}}{2}\right), \quad y=\frac{1}{2 \lambda} y_{1},  \tag{A.16}\\
z=\frac{1}{\lambda^{2}}\left(-x_{1}+2 \delta^{2} z_{1}\right) .
\end{gather*}
$$

Using (A.15) and (A.16) we can finally rewrite the set of equations in the canonical form:

$$
\begin{gather*}
\dot{x_{1}}=-\lambda y_{1}+\frac{\delta \gamma}{2 \lambda^{3}} y_{1} x_{1}-\frac{\gamma \delta^{3}}{\lambda^{3}} y_{1} z_{1} \\
\dot{y_{1}}=\lambda x_{1}+\frac{\gamma \delta\left(\delta^{2}-1\right)}{\lambda^{3}} x_{1} z_{1}-\frac{\gamma \delta}{2 \lambda^{3}} x_{1}^{2}+\frac{2 \gamma \delta^{3}}{\lambda^{3}} z_{1}^{2}  \tag{A.17}\\
\dot{z_{1}}=\frac{\gamma}{4 \lambda^{3} \delta} y_{1} x_{1}-\frac{\gamma \delta}{2 \lambda^{3}} y_{1} z_{1} .
\end{gather*}
$$

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