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Observation of phase synchronization and alignment during free induction decay of quantum spins with Heisenberg interactions

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Abstract

Equilibration of observables in closed quantum systems that are described by a unitary time evolution is a meanwhile well-established phenomenon apart from a few equally well-established exceptions. Here we report the surprising theoretical observation that integrable as well as non-integrable spin rings with nearest-neighbor or long-range isotropic Heisenberg interaction not only equilibrate but moreover also synchronize the directions of the expectation values of the individual spins. We highlight that this differs from spontaneous synchronization in quantum dissipative systems. Here, we observe mutual synchronization of local spin directions in closed systems under unitary time evolution. In our numerical simulations, we investigate the free induction decay of an ensemble of up to $N = 25$ quantum spins with $s = 1/2$ each by solving the time-dependent Schrödinger equation numerically exactly. Our findings are related to, but not fully explained by conservation laws of the system. Even if we cannot provide a full understanding of the phenomenon, it is very robust against for instance random fluctuations of the Heisenberg couplings and inhomogeneous magnetic fields. The observed synchronization is independent of whether the interaction is ferro- or antiferromagnetic. Synchronization is not observed with strong enough symmetry-breaking interactions such as the dipolar interaction. We also compare our results to closed-system classical spin dynamics which does not exhibit phase synchronization due to the lack of entanglement and since the fixed magnitude of individual classical spins effectively acts like additional *N* conservation laws.

1. Introduction

Decoherence, equilibration as well as thermalization in closed quantum systems under unitary time evolution are well-studied and by now well-established concepts which root in seminal papers by Deutsch, Srednicki and many others $[1-12]$ $[1-12]$. For numerical studies, spin systems are the models of choice both since they are numerically feasible due to the finite size of their Hilbert spaces as well as they are experimentally accessible for instance in standard investigations by means of electron parametric resonance, free induction decay (FID), or in atomic traps, see e.g. $[13-17]$ $[13-17]$. In such systems, observables assume expectation values that are practically indistinguishable from the prediction of the diagonal ensemble for the vast majority of all times of their time evolution $[6, 18]$ $[6, 18]$ $[6, 18]$.

In this paper we provide a first discussion of an observation that rests both on decoherence and equilibration. We study the FID of quantum spins that are arranged on a ring-like geometry with nearest-neighbor as well as long-range isotropic Heisenberg interactions. For the overwhelming majority of investigated cases the initial product state of single-spin states entangles, i.e. turns into a superposition of product states, and thereby equilibrates at the level of single-spin observables. Our most striking observation is that expectation values of all individual spin vectors synchronize with respect to their orientation. In an FID setting this means that their various individual rotations about the common field axis synchronize and align in the course of time. In a co-rotating frame they simply align. Experimentally, such collective effects may e.g. be imprinted in the temporal line shapes of the optical response under ultrashort pulse excitation and thus eventually be observed [\[19\]](#page-18-6).

We would like to contrast our findings with the longer-known observation of (spontaneous) synchronization in dissipative systems [\[20](#page-18-7)[–24\]](#page-18-8). It was controversially discussed whether quantum two-level systems are able to synchronize at all [\[25\]](#page-18-9), with later conclusions that this is indeed the case [\[22,](#page-18-10) [26\]](#page-18-11). Stable limit cycles of the participating oscillators play an important role in several systems, but there are also other mechanisms at work. Since such a discussion is applicable only to dissipative systems, which can emit or absorb energy to return to their stable oscillation after a perturbation, it probably cannot serve as an explanation in our case.

A related and already investigated topic is transient synchronization in open quantum systems [\[27\]](#page-18-12), in which the system finally equilibrates to a non-synchronized state, but synchronizes temporarily on the way. We show that we observe a comparable behavior in closed quantum spin systems in the form of a pre-equilibration phenomenon, if we weakly reduce the symmetry of the Hamiltonian. Similar to the known cases of transient synchronization, the weaker the perturbation (in our case symmetry breaking interactions) the longer this pre-equilibration regime lasts.

The observed synchronization is stable against random fluctuations of the Heisenberg couplings and we observe it for almost all initial conditions. We therefore conjecture that it is tightly connected to the symmetries and conserved quantities of the isotropic Heisenberg model which is SU(2) invariant [\[28\]](#page-18-13), see also [\[29\]](#page-18-14). This hypothesis is corroborated by the observation that strongly anisotropic interactions such as the dipolar interaction spoil the synchronization. Also in classical spin dynamics the phenomenon cannot be observed as will be discussed in detail later. Inhomogeneous or randomly fluctuating local fields at the sites of the individual spins on the other hand do not prevent the spins from synchronizing although several conservation laws are broken. The same applies for weakly anisotropic interactions that are close to the isotropic Heisenberg case; here we observe a transient synchronization.

The paper is organized as follows. In section [2](#page-2-0) we introduce the theoretical model and the applied methods. Section [3](#page-4-0) deals with exemplary numerical quantum simulations under isotropic Heisenberg interactions and we compare to classical simulations. Section [4](#page-8-0) introduces symmetry breaking anisotropic interactions and demonstrates the transient behavior of the synchronization phenomenon. Section [5](#page-10-0) provides a summary of our main results. In the appendix some aspects are discussed in more detail, especially the behavior under symmetry breaking interactions. Video clips of our simulations are provided on the website of the paper [\[30\]](#page-18-15).

2. Theoretical model and methods

The Hamiltonian of our spin model reads

$$
H = -\sum_{j=1}^{N} J_j \vec{s}_j \cdot \vec{s}_{j+1} - \sum_{j=1}^{N} h_j \vec{s}_j, \tag{1}
$$

where the first sum corresponds to the isotropic Heisenberg model and the second sum denotes the Zeeman term. Operators are marked by a tilde, the Heisenberg interactions are denoted by *Jj*, local magnetic fields are given by h_j , and periodic boundary conditions $\vec{g}_{N+1} = \vec{g}_1$ are applied. Thus, the Hamiltonian describes \sim ^{*N*T₁ \sim} spins which are arranged as a ring; it could for instance be a ring molecule [\[31,](#page-18-16) [32\]](#page-18-17). We define the total spin operator

$$
\vec{\mathbf{S}} := \sum_{k=1}^{N} \vec{\mathbf{S}}_k,\tag{2}
$$

which commutes with the Heisenberg part of the Hamiltonian, and so does \vec{S} \tilde{a} ², even if the coupling constants J_i are all different. This is true for any spin arrangement, not just for rings [\[28,](#page-18-13) [33\]](#page-18-18). The

conservation of \vec{S}^2 is broken either by anisotropic interactions or by varying local magnetic fields h_j \tilde{a}

$$
\left[\vec{S}^2, \sum_{i=1}^N h_{i\vec{S}_i}^z\right] \propto \left[\vec{S}_i \cdot \vec{S}_j, h_{i\vec{S}_i}^z + h_{j\vec{S}_j}^z\right].
$$
\n(3)\n
\n= 0 only if $h_i = h_j$

Furthermore, we define the transverse magnetization

$$
M_{\text{trans}} := \sqrt{\langle \underline{S}^x \rangle^2 + \langle \underline{S}^y \rangle^2}
$$

=
$$
\sqrt{\left(\sum_j \langle \underline{S}^x_j \rangle\right)^2 + \left(\sum_j \langle \underline{S}^y_j \rangle\right)^2}.
$$
 (4)

Here $\langle S^x \rangle$ denotes the expectation value with respect to a specified many-body state. We interpret [\(4\)](#page-3-0) as the ∼ net magnetization precessing in the *xy*-plane. In case of Hamiltonian [\(1\)](#page-2-1) this is also a conserved quantity if the local magnetic fields are all the same $h_j \equiv h \ \forall j$. This can be seen by looking at the time evolution $(\hbar := 1)$

$$
\frac{d}{dt}\langle\psi(t)|\vec{S}|\psi(t)\rangle = \frac{1}{i}\langle\psi(t)|[\vec{S},H]|\psi(t)\rangle
$$

= $i\hbar\langle\psi(t)|[\vec{S},S^z]|\psi(t)\rangle$. (5)

Remember, *h* denotes the magnetic field. The solution of equation [\(5\)](#page-3-1) is of the form

$$
\langle \psi(t) | \vec{\mathbf{S}} | \psi(t) \rangle = \begin{pmatrix} a \cos ht + b \sin ht \\ -b \cos ht + a \sin ht \\ c \end{pmatrix} , \qquad (6)
$$

compatible with the conserved quantities. The coefficients *a*, *b* and *c* are determined by the initial state of the system. We observe a collective rotation with frequency *h* in all cases the spins synchronize (sections [3.1,](#page-4-1) [3.2](#page-4-2) and [3.4\)](#page-7-0). Appendix [B.1](#page-14-0) provides an exception where the spins collectively precess around a mean field \tilde{h} .

As initial many-body states we choose product states of the form

$$
|\psi(t=0)\rangle = \bigotimes_{j=1}^{N} \frac{1}{\sqrt{2}} (|\uparrow\rangle + e^{i\theta_{j}}|\downarrow\rangle), \qquad (7)
$$

for which the expectation values of individual spins

$$
\langle \vec{\mathbf{z}}_j \rangle := \langle \psi | \vec{\mathbf{z}}_j | \psi \rangle \tag{8}
$$

are oriented in the *xy*-plane and point in a direction that depends on θ_j . In the following we are going to investigate the time evolution of the four states shown in figure [1](#page-4-3) where (a) all spins point in the same direction, (b) are regularly fanned out by 180 degrees, (c) are regularly fanned out by 360 degrees, and (d) point in random directions. We will refer to these states as $|\psi_A\rangle$, $|\psi_B\rangle$, $|\psi_C\rangle$, and $|\psi_D\rangle$ (or A, B, C, and D in the classical case, section [3.5\)](#page-7-1).

In a product state, the spins are not entangled by definition, however they entangle during the unitary time evolution

$$
|\psi(t)\rangle = e^{-iHt} |\psi(0)\rangle, \tag{9}
$$

that we calculate numerically exactly using a Suzuki Trotter decomposition [\[34\]](#page-18-19).

In order to measure the entanglement of an individual spin at site *j* with the others, we define the reduced density matrix

$$
\varrho_j = \text{Tr}_{\bigotimes_{k \neq j} \mathscr{H}_k} \left(|\psi\rangle\langle\psi| \right). \tag{10}
$$

Here \mathscr{H}_j denotes the Hilbert subspace of spin *j*, and $\mathscr{H}=\bigotimes_{j=1}^N \mathscr{H}_j$ is the total Hilbert space. The purity is given as $Tr(\rho_j^2)$. $Tr(\rho_j^2) = 1$ holds, if spin *j* is not entangled with other spins, and $Tr(\rho_j^2) = 0.5$ if it ˜ ˜ ˜ is maximally entangled with other spins. The purity is thus also a measure of decoherence for an observer of

a single spin [\[35,](#page-18-20) [36\]](#page-18-21). An alternative way of quantifying the decoherence would be the von Neumann entropy $S(\rho_j) = -Tr(\rho_j \log_2 \rho_j)$ [\[37\]](#page-18-22). \tilde{a} \tilde{a} \tilde{a}

3. Calculations and results

In this section we present our numerical findings of the special behavior of initial states in figure [1](#page-4-3) under time evolution with Hamiltonian equation [\(1\)](#page-2-1) and equal magnetic fields $h_j = -1 \forall j$. As discussed, M_{trans} and \overline{S} \tilde{a} 2 are conserved quantities. We show that, with one exception, the spin expectation values synchronize.

3.1. Initial state $|\psi_{A}\rangle$

In figure [2](#page-4-4) we start with initial state $|\psi_A\rangle$ and random Heisenberg interactions *J_i*. In this case, every spin is precessing as if independent without entangling to other spins, no matter how the *Jj* are chosen. Since all spins point in the same direction, M_{trans} and \vec{S}^2 assume their maximum values. Because they are conserved $\tilde{}$ quantities the spins are bound to remain in a perfect product state, otherwise it would not be possible to conserve these values over time.

Figure 3. Time evolution of initial state $|\psi_B\rangle$ w.r.t. Hamiltonian equation [\(1\)](#page-2-1) with isotropic Heisenberg interactions and *J_j* ∈ [1.6, 2.4], *h_i* = −1 $\forall j$, *N* = 25. The video for (a) [\(https://stacks.iop.org/NJP/23/083038/mmedia\)](https://stacks.iop.org/NJP/23/083038/mmedia) are provided in the supplementary data.

3.2. Initial state $|\psi_{\text{B}}\rangle$

Figure [3](#page-5-0) shows almost the same as figure [2,](#page-4-4) but this time for initial state $|\psi_B\rangle$. Initially the individual spin expectation values are spread out by 180 degrees, but during time evolution they align. This astonishing phenomenon can be nicely observed in the video provided on the web page of the published article [\[30\]](#page-18-15). During time evolution and synchronization the spins entangle as much as the conservation of \vec{S}^2 and

*M*_{trans} allows. Interestingly, the spins stay entangled and do not fan out again (apart from finite size effects such as revivals at very late times). This statement becomes stronger with increasing system size, which is further addressed in appendix [A.](#page-11-0) We interpret this phenomenon as quantum mechanical equilibration process under the restricting influence of conserved quantities [\[29\]](#page-18-14).

The synchronization can be rationalized for spin systems where all spins are equivalent, i.e. ring systems with translational invariance $(J_i = J, h_j = h \forall j)$ since then equilibration should result in the same single-spin expectation value at every site. This concerns magnitude and direction of the spin vector. The somewhat unexpected result of our investigation is that the direction of all spins continues to synchronize also for settings where spins are no longer equivalent, i.e. if the Heisenberg interactions are drawn at random from a distribution.

Figure [3\(](#page-5-0)c) shows the purity of the individual reduced density operators ρ_j (equation [\(10\)](#page-3-3)). Since the

 \tilde{a} couplings *Jj* are different for different *j*, not all spins are equal. This does not prevent the spins from synchronizing their directions, but they do not all entangle to the same extent.

J_j ∈ [1.6, 2.4], *N* = 24 without (a) and (b) and with magnetic field (c) and (d). Video (c) is provided in the supplementary data.

Another main result of this paper is that the time needed for the spins to synchronize is almost independent of the width Δ of the distribution of the *J_i* ∈ [2 − Δ , 2 + Δ]. This is also demonstrated numerically in appendix [A.](#page-11-0)

Figure [4](#page-5-1) shows the variance of the expectation values of individual spin operators, defined as

$$
\text{Var}(\langle \zeta_j^x \rangle)(t) := \frac{1}{N} \sum_{j=1}^N \left(\langle \zeta_j^x \rangle - \frac{\langle S^x \rangle}{N} \right)^2 \tag{11}
$$

for different system sizes *N*. That the variance decays to zero, compare figure [4,](#page-5-1) expresses precisely that the spins align until they point in the same direction. This process takes the longer the larger the system is. The synchronisation, i.e. the alignment of directions, also takes place in the absence of a magnetic field, as can be seen in figure [4\(](#page-5-1)a). The reason is that the homogeneous magnetic field, which is a one-body

operator, does not causes any many-body entanglement between the spins; entanglement and equilibration are driven by the Heisenberg term which is a two-body operator. As a result, the field-free curves in figure $4(a)$ $4(a)$ are the envelopes of the curves taken with homogeneous field and shown in figure $4(b)$.

3.3. Initial state $|\psi_{\text{C}}\rangle$

Figure [5](#page-6-0) shows the time evolution for initial state $|\psi_C\rangle$ and different system sizes. This is a very special and atypical case with a particular symmetry in the spin orientations which results in a very stable state even if there are different couplings J_i between the spins, see video [\[30\]](#page-18-15). This is the only initial state we find where the spins do not align, but entangle and decay to zero, with wild echos at later times. The larger the system, the longer the echos take to occur and the longer it takes for the spins to entangle.

These numerical results for finite system sizes suggest that $|\psi_C\rangle$ is an energy eigenstate in the thermodynamic limit which appears plausible, because the angle between neighboring spins is given by $2\pi/N$, therefore for $N \to \infty$ all neighbors are parallel in the initial state. We emphasize that this state would also be an energy eigenstate in the non-integrable case [\[38,](#page-18-23) [39\]](#page-18-24) where the *Jj* are all different. Because all single-spin observables are strongly different we conjecture that this state is not thermal; its relation to quantum scars needs to be explored, see [\[40,](#page-18-25) [41\]](#page-18-26) and references therein.

3.4. Initial state $|\psi_{\text{D}}\rangle$

Figure [6](#page-6-1) shows a time evolution for initial state $|\psi_D\rangle$ (random orientations) without magnetic field (figure $6(a)$ $6(a)$) and with magnetic field (figure $6(b)$). The conserved net magnetization is small (would be zero in the thermodynamic limit or as a mean of many random realizations according to the central limit theorem). Nevertheless, the spins synchronize which shows that this phenomenon is very robust with respect to the initial state, see also video [\[30\]](#page-18-15).

3.5. Classical spin dynamics

Although not at the heart of our investigations, we like to compare our results to classical spin dynamics with identical Heisenberg couplings and initial states. It turns out that a classical spin dynamics does not exhibit phase synchronization, see videos [\[30\]](#page-18-15). The reason in this context is that classical spin dynamics lacks entanglement which is necessary for synchronization. Contrary to the expectation values of the quantum spins, the classical spins are bound to maintain their length, which effectively acts like additional *N* conservation laws. This results in an oscillatory dynamics of most investigated initial conditions, compare figure [7](#page-7-2) for initial states equivalent to figures $2(b)$ $2(b)$ –(d). Initial state C is again special. In the quantum case the spins maintain their directions for a long time while slowly entangling. In the classical case the spins also keep their directions for a long time. This is because of the high symmetry in the initial state with zero net magnetization and no preferred direction in the *xy*-plane.

The classical spin dynamics has been evaluated according to

$$
\frac{\mathrm{d}}{\mathrm{d}t}\vec{s}_j = \frac{\partial H}{\partial \vec{s}_j} \times \vec{s}_j,\tag{12}
$$

where *H* denotes the classical Hamiltonian analogous to equation [\(1\)](#page-2-1).

In the literature one finds well-studied classical examples of synchronization of e.g. Van der Pol oscillators [\[42\]](#page-19-0). These are dissipative systems with stable limit cycles, which means that after a perturbation they return to their ordinary oscillation. This would not be the case for our classical spins even if we would couple them to a heat bath.

One trivial (and much different) way of how classical spins would synchronize (at least in a transient way) is by choosing e.g. ferromagnetic (or antiferromagnetic) Heisenberg couplings *Jj* and a dissipative dynamics so that the ferromagnetic (or antiferromagnetic) ground state is approached, which is aligned trivially.

Figure [8](#page-8-3) shows such an example for different intervals of the Heisenberg coupling strength. The damping is realized by

$$
\frac{\mathrm{d}}{\mathrm{d}t}\vec{s}_j = \frac{\partial H}{\partial \vec{s}_j} \times \vec{s}_j - \alpha \left(\frac{\partial H}{\partial \vec{s}_j} \times \vec{s}_j\right) \times \vec{s}_j. \tag{13}
$$

Because of the damping, the system looses energy until it arrives in the lowest possible energy state. For a ferromagnetic coupling the spins synchronize in a parallel fashion, for a bipartite antiferromagnetic system the spins synchronize in an antiparallel fashion. In figure [8](#page-8-3) there is also a magnetic field applied in *z*-direction with which the spins also align. Therefore, at late times all spins point in *z*-direction. If the ferromagnetic coupling is much larger than the magnetic field $|J_i| \gg |h|$, then also the *x*- and *y*-components of the spins synchronize in a transient way.

We note that such kind of synchronization is fundamentally different from the synchronization we observe in closed quantum spin systems. In the quantum case, it does not matter if the Heisenberg interaction is ferro- or antiferromagnetic, the single-spin vector observables always synchronize in a parallel fashion. The synchronization is just based on equilibration, entanglement, and conservation laws.

4. Breaking the symmetry

In this section we investigate whether synchronization still occurs if M_{trans} and \vec{S}^2 are not conserved ∼ anymore. We can break the symmetry in different ways, either by means of inhomogeneous magnetic fields

or by interactions between the spins that are not of isotropic Heisenberg type. In section [4.1](#page-9-1) we choose XYZ interactions and in section [4.2](#page-10-2) XX interactions as two examples with different outcomes. In appendix [B](#page-14-1) we show the effect of inhomogeneous magnetic fields (appendix [B.1\)](#page-14-0) and of dipolar interactions between all spins (appendix [B.2\)](#page-16-0).

4.1. XYZ interaction

We begin with the XYZ interaction which is close to the isotropic Heisenberg case if the interaction in the three spatial directions is not too different. In this case, the synchronization between the spins still occurs. The Hamiltonian in this subsection is defined as

$$
H_{\sim XYZ} = -J \sum_{j=1}^{N} \tilde{s}_j^x \tilde{s}_{j+1}^x - (J - \delta) \sum_{j=1}^{N} \tilde{s}_j^y \tilde{s}_{j+1}^y - (J - 2\delta) \sum_{j=1}^{N} \tilde{s}_j^z \tilde{s}_{j+1}^z - h \sum_{j=1}^{N} \tilde{s}_j^z.
$$
 (14)

We use the parameter δ to tune the difference of the interaction in the three spatial directions.

Figure [9](#page-9-2) shows time evolutions for initial state $|\psi_B\rangle$ and two different values of δ . The magnetization is not a conserved quantity anymore and will therefore decay toward its equilibrium value, which is zero in the *xy*-plane for a magnetic field in *z*-direction. Our investigations reveal that the larger δ the faster the spins decay. However, we clearly observe that while decaying the spins still synchronize, see especially figure [9\(](#page-9-2)a). One could say, that the synchronization is a transient phenomenon in such cases since the time scale of synchronization is shorter than that of the unavoidable decay.

Figures [9\(](#page-9-2)b) and (d) show the purity of the reduced density operators ρ_j introduced in equation [\(10\)](#page-3-3). We see that all spins maximally entangle ($Tr(\rho_j^2) = 0.5$) which is equivalent with all individual spin \tilde{a} expectation values decaying to zero.

Another example of broken symmetry where the spins still synchronize is shown in appendix [B.1](#page-14-0) with an inhomogeneous magnetic field.

4.2. XX interaction

As comparison we now show a case with XX interaction where the spins do not synchronize. The Hamiltonian is defined as

$$
H_{\sim XX} = -J \sum_{j=1}^{N} \left(g_j^x g_{j+1}^x + g_j^y g_{j+1}^y \right) - h \sum_{j=1}^{N} g_j^z.
$$
 (15)

Figure [10](#page-9-3) shows a time evolution for initial state $|\psi_B\rangle$. The decay of the transverse magnetization is much faster than in the previous subsection, because we are further away from isotropic Heisenberg interactions and the symmetry regarding the conservation of the transverse magnetization is broken much more strongly, compare also [\[17\]](#page-18-3). In order to see if the spins still synchronize while decaying to zero, we choose a much smaller coupling constant $J = 0.1$ instead of $J = 2$. But we clearly see that the spins do not synchronize while decaying or equivalently the timescale of the decay is much shorter than the timescale of synchronization.

Another example of broken symmetry where the spins do not synchronize is shown in appendix [B.2](#page-16-0) with dipolar interactions between the spins.

In a future investigation, we plan to study the relation of our findings for the translationally invariant XXZ case with the suggestion of a generalized Gibbs ensemble as the long-time limit of the unitary dynamics induced by dynamical symmetries according to references [\[43–](#page-19-1)[45\]](#page-19-2).

5. Summary

As a conclusion we can say first of all that the conservation of M_{trans} and \vec{S}^2 not just slows down the FID, ∼ but prevents the free induction from decaying if the Hamiltonian only contains isotropic Heisenberg interactions and the Zeeman terms of all spins are equal (see figure [2\)](#page-4-4). This is in accord with reference [\[29\]](#page-18-14).

Furthermore, we demonstrate in detail the interesting phenomenon that the single-spin vector expectation values align in the course of time almost independent of how they are initialized in the *xy*-plane (see figure [3\)](#page-5-0). It does not matter if the initial state is a product state of the form equation (7) or if the spins start in an entangled state (see appendix [A.3\)](#page-12-0). For the process of synchronization the magnetic field is not necessary, it only induces a (collective) rotation of all spins about the field axis (see e.g. figures [4](#page-5-1) and [11\)](#page-12-1). The Heisenberg interactions cause an equilibration process under the constraint of conserved quantities. In the case of SU(2) symmetry combined with translational symmetry and under the assumption of equilibration (of one-body observables) all single-spin expectation values must assume the same direction and magnitude. Translational invariance requires that each single-spin observable is the same. We show that after entanglement is maximised (under constraints of conserved quantities) and equilibration is completed the spins stay synchronized and fluctuate the less the larger the system is (see e.g. figure [14,](#page-13-0) appendix [A.1\)](#page-11-1). However, we demonstrate that synchronization also occurs without translational symmetry and that the timescale of synchronization is independent of the width Δ of the distribution of Heisenberg couplings J_i (see figure [15,](#page-14-2) appendix [A.1\)](#page-11-1).

We demonstrate that such a synchronization is not possible with classical spins in a closed system (see figure [7\)](#page-7-2). Moreover we give an example for dissipative classical spins which experience a transient synchronization based on ferromagnetism (see figure [8\)](#page-8-3). We highlight that this is very different from the observed quantum mechanical synchronization where the system is closed and the sign of the Heisenberg interaction does not matter—the single-spin vector observables always synchronize in a parallel fashion along the direction of the total spin.

In addition, we discuss that the synchronization of spin expectation values is very robust. It happens already for small systems ($N = 10$, see figure [13,](#page-13-1) appendix [A\)](#page-11-0) and for various initial states (see e.g. figure [6\)](#page-6-1). We find just one exception (initial state $|\psi_c\rangle$) which has a special symmetry and in the thermodynamic limit becomes an energy eigenstate (see figure [5\)](#page-6-0). Synchronization also occurs for long-range Heisenberg interactions (see figure [21,](#page-17-0) appendix [B.2\)](#page-16-0). Synchronization is not limited to spins $s = 1/2$. In appendix [A.2](#page-11-2) we demonstrate that also spins with $s = 1$ synchronize under isotropic Heisenberg interactions.

Further on, we provide examples of transient synchronization for systems where symmetries are broken, because the time scale of synchronization is shorter than that of equilibration. Systems with anisotropic XYZ interactions belong to this set if they are still close to the isotropic Heisenberg case (see figure [9\)](#page-9-2), or if the symmetry is broken by means of an inhomogeneous magnetic field (see figure 18 and appendix $B.1$, respectively).

Finally, we show that spins do not synchronize for interactions that are strongly anisotropic such as dipolar interactions (see figure [20,](#page-17-1) appendix [B.2\)](#page-16-0).

Our investigations might be helpful for interpreting observations in the context of FID. Even if we cannot provide a complete analytical explanation and interpretation of the phenomenon, we think the wide range of numerical results demonstrates that the phenomenon of synchronization is widespread and robust.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A. Additions to section [3](#page-4-0)

A.1. More data regarding section [3.2](#page-4-2)

Here we present more detailed numerical calculations regarding section [3.2.](#page-4-2) In figure [11](#page-12-1) the purity of individual reduced density matrices is shown for different system sizes *N*. All Heisenberg couplings *J_i* are chosen equal and therefore all spins entangle in an equal way during equilibration (in contrast to figure $3(c)$ $3(c)$).

It can be clearly seen that the respective purities fluctuate less the larger the system is. Thus, in the thermodynamic limit $(N \to \infty)$ we expect them to keep the same order of magnitude without fluctuating after equilibration. This figure is completely independent of the strength of the magnetic field *h*, because the Zeeman term in Hamiltonian equation [\(1\)](#page-2-1) does not cause any entanglement between spins.

Figure [12](#page-12-2) shows the individual $\langle s_j^z \rangle$ expectation values for exactly the same time evolutions as figure [11.](#page-12-1) Initially all these values are zero because the spins are oriented in the *xy*-plane (see figure [1\)](#page-4-3). During time evolution (especially at the beginning) the spins leave the *xy*-plane, but at later times these fluctuations in *z*-direction become small for a larger system size *N*.

Figures [13](#page-13-1) and [14](#page-13-0) show individual spin expectation values for time evolutions without magnetic field for different system sizes and for short and long time (also for initial state $|\psi_{\rm B}\rangle$). It can be qualitatively seen that such synchronization of spins can already be observed for a small system size of $N = 10$. For $N = 6$ the spins fluctuate much and for $N = 2$ the spins permanently point in opposite directions.

We now want to focus on the question how the choice of couplings J_i influences the time needed for the spins to align. This is addressed by figure [15.](#page-14-2) The couplings J_i are chosen randomly from intervals of different width Δ and the magnetic field is zero. We see that Δ has a very small impact on the time evolution and the behavior of the spins; the process of alignment and the time it takes is very robust. The time to synchronization does only depend on the system size *N* and the mean *Jj*.

A.2. Spins $\zeta = 1$

The question arises if the observed synchronization phenomenon also occurs with larger spins than $s = 1/2$. To address this question at least partly, we simulate spin rings with spin quantum numbers $\zeta = 1$. To simplify our numerical calculation we form these spins by coupling two spins with $s = 1/2$ each ferromagnetically. We define

$$
\vec{\zeta}_j := \vec{\zeta}_{2j} + \vec{\zeta}_{2j-1}.
$$
\n(A1)

Instead of *N* spins with $s = 1/2$ we have $\tilde{N} = N/2$ spins with $\zeta = 1$. The following Hamiltonian applies

$$
H = -\sum_{j=1}^{\tilde{N}} J_j \vec{\zeta}_j \cdot \vec{\zeta}_{j+1} - \sum_{j=1}^{\tilde{N}} h_j \zeta_j^z - J_f \sum_{j=1}^{\tilde{N}} \vec{\xi}_{2j} \cdot \vec{\xi}_{2j-1},
$$
 (A2)

The last term in equation [\(A2\)](#page-11-3) is the strong ferromagnetic coupling, $J_f = 100 \gg |J_i|$, between every two

fused spins. We initialize the state of the system such that those two coupled spins point in the same direction and the \tilde{N} spins are fanned out by 180 degrees. The time evolution is shown in figure [16,](#page-14-3) and indeed the spins still synchronize.

We thus speculate that there is no obvious or simple classical limit by which synchronizations would disappear when going from small spin quantum numbers to large spin quantum numbers. Also the time scale on which synchronization happens does not seem to be very different between spins $s = 1/2$ and $\zeta = 1$, compare figures [16\(](#page-14-3)a) and [13\(](#page-13-1)d). This question certainly needs further investigations.

A.3. Starting in an entangled state

So far our initial state was always a product state equation [\(7\)](#page-3-2). That might give the impression that this could be relevant for the phenomenon of synchronization. But this is not the case, as we show with one example in this subsection.

Consider the state

$$
|\phi\rangle = \sum_{k=1}^{N} c_k |k\rangle, \tag{A3}
$$

with Gaussian distributed random coefficients and an arbitrary basis $|k\rangle$ of the Hilbert space. Such a state will be maximally entangled and all spin expectation values are close to zero. From this we construct our

initial state

$$
|\psi\rangle \propto \left(\prod_{j=1}^{N} e^{\epsilon \sin(\alpha_j) \underline{s}_j^x} e^{\epsilon \cos(\alpha_j) \underline{s}_j^y}\right) |\phi\rangle, \tag{A4}
$$

with $\alpha_j = \frac{j\pi}{N}$ and ϵ a parameter for the magnitude of the deflection of the spin expectation values from zero. This way we create a (still entangled) state with single spin expectations values $\neq 0$ in the *xy*-plane fanned out by 180 degrees. Figure [17](#page-15-1) shows associated time evolutions and that the spins do indeed still synchronize, starting in an entangled state equation [\(A4\)](#page-14-4).

Appendix B. Additions to section [4](#page-8-0)

In addition to section [4](#page-8-0) we present more cases of how the spins behave with broken symmetry (without the conserved quantities M_{trans} and \vec{S} \tilde{a} 2).

B.1. Inhomogeneous magnetic field

As shown in equation [\(3\)](#page-3-4) all magnetic fields *hj* have to be equal or the conserved quantities are broken. Figure [18](#page-15-0) shows time evolutions of initial state $|\psi_{\rm B}\rangle$ where only a few spins see a magnetic field $h_i = h \neq 0$ and all others fields are zero. Surprisingly the spins do still synchronize and precess together while they decay. The spins without magnetic field are carried with the others.

The frequency \tilde{h} of the collective precession decreases the more spins there are with $h_j = 0$. This can be viewed as every spin sees a mean field $\tilde{h} = \sum_j h_j/N$. From figures [18\(](#page-15-0)a)–(e) the number of magnetic fields $h_j = h$ is halved and the precession frequency \tilde{h} also halves.

Another way of breaking the symmetry is by choosing random magnetic fields. Figure [19](#page-16-1) shows time evolutions for initial state $|\psi_B\rangle$ where the magnetic fields h_j are drawn at random from a distribution of different width ξ. The spins do still synchronize up to large values of ξ up to the point where the decay of the transverse magnetization is faster than the synchronization.

B.2. Dipolar and long range interactions

We now investigate how initial state $|\psi_B\rangle$ behaves when all spins (not only neighbors) interact with dipolar interactions or just long-range Heisenberg interactions.

The Hamiltonian of a dipolar interacting spin system is given by

$$
H = \sum_{j=1}^{N} \sum_{k=j+1}^{N} \frac{\lambda}{r_{jk}^3} \left(\vec{\xi}_{j} \cdot \vec{\xi}_{k} - \frac{3(\vec{\xi}_{j} \cdot \vec{r}_{jk})(\vec{\xi}_{k} \cdot \vec{r}_{jk})}{r_{jk}^2} \right) - h \sum_{j=1}^{N} \vec{\xi}_{j}^z.
$$
(B1)

We use the parameter λ to tune the strength of the interaction. We also need spatial coordinates \vec{r}_j for all spins. For simplicity we arrange them on the unit circle

$$
\vec{r}_j = \begin{pmatrix} \cos(2\pi j/N) \\ \sin(2\pi j/N) \\ 0 \end{pmatrix} . \tag{B2}
$$

Figure [20](#page-17-1) shows time evolutions for initial state $|\psi_B\rangle$ for two different parameters λ . As expected the spin expectation values decay the faster the larger the parameter λ is. But even for a small λ in figure [20\(](#page-17-1)a) the spins do not synchronize at all. This can also be seen in the related video at [\[30\]](#page-18-15). Dipolar interactions are highly anisotropic, therefore, the conservation of the transverse magnetization is broken strongly.

Finally, we want to test if synchronization under isotropic Heisenberg couplings is limited to nearest-neighbor interactions or holds under long-range interactions. To this end, we take as an example the above Hamiltonian equation $(B1)$ and remove the anisotropic parts

$$
H = \sum_{j=1}^{N} \sum_{k=j+1}^{N} \frac{\lambda}{r_{jk}^3} \vec{z}_j \cdot \vec{z}_k - h \sum_{j=1}^{N} \vec{z}_j^z.
$$
 (B3)

Again we tune the interaction strength by the parameter λ . Figure [21](#page-17-0) shows calculations for very small to larger values of λ . For $\lambda = 0.005$ in figure [21](#page-17-0) the time scale shown is not sufficient for the spins to

synchronize, whereas in figure $20(c)$ $20(c)$ there is enough time for the magnetization to decay completely. This shows that the magnetization decay through the anisotropic terms of the dipolar interaction happens on a much faster time scale than the isotropic part causes synchronization. Choosing λ significantly larger in figure [21](#page-17-0) leads to synchronization in the given time frame.

One finding of this chapter is that the synchronization effect is more general, and it does not only appear for nearest-neighbor isotropic Heisenberg interactions.

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