Experimental demonstration of a stimulated polarization wave in a chain of nuclear spins

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Abstract. A stimulated wave of polarization, which implements a simple mechanism of quantum amplification, is experimentally demonstrated in a chain of four $J$-coupled nuclear spins, irradiated by a weak radio-frequency transverse field. The ‘quantum domino’ dynamics, a wave of flipped spins triggered by a flip of the first spin, has been observed in fully $^{13}$C-labelled sodium butyrate.

In quantum dynamics, governed by linear equations of motion, amplification can be realized by using entangled quantum states of a system. In this case, a local perturbation, affecting a small part of the system, changes a wavefunction of the entire system in a coherent way. Subsequently, this change may be converted by dynamic evolution into changes of ‘macroscopic’ observables [1]–[3]. Instead of this two-step approach, one can incorporate the entangling operations into a single dynamical process. For an $N$-qubit system, one of the simple logical schemes of this process is a chain of unitary controlled-not operations [3]

$$U = \text{CNOT}_{N-1,N}\text{CNOT}_{N-2,N-1}\cdots\text{CNOT}_2,3\text{CNOT}_{1,2},$$

(1)

where CNOT$_{m,n}$ flips the state of the $n$th qubit if the $m$th qubit is in the state $|1\rangle$ and does not do anything if the state is $|0\rangle$. The unitary transformation (1) converts the initial state

$$|\psi_m\rangle = (a|0\rangle_1 + b|1\rangle_1)|0\rangle_2|0\rangle_3\cdots|0\rangle_{N-1}|0\rangle_N$$

(2)

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into the final state

$$|\psi_{\text{out}}\rangle = U |\psi_{\text{in}}\rangle = a|0\rangle_1|0\rangle_2 \cdots |0\rangle_{N-1}|0\rangle_N + b|1\rangle_1|1\rangle_2 \cdots |1\rangle_{N-1}|1\rangle_N,$$

(3)

where the quantum state of the first qubit is ‘expanded’, and the state of polarization of this single qubit is transferred to the state of the total polarization of the entire cluster. One can see that the chain of gates (1) offers an efficient way to create entanglements. At $a = b = 2^{-1/2}$ the output state (3) is the maximally entangled ‘Schrödinger cat’ state.

If viewed as consecutive steps in time, one gate after another, the chain (1) describes a ‘quantum domino’ dynamics, when a wave of flipped qubits is triggered by a flip of the first qubit. It is also possible to create schemes where the number of flipped spins grows faster than linear with the number of logic steps [4]. A physical model with continuous dynamics, similar to that suggested by the chain (1), is an Ising chain with nearest-neighbour interactions, irradiated by a weak resonant transverse field [5]. The Hamiltonian of this model is

$$H = \frac{\omega_0}{2} \sum_{i=1}^{N} \sigma_z^i + \omega_1 \sum_{i=1}^{N} \sigma_z^i \cos \omega_0 t + \frac{J}{4} \sum_{i=1}^{N-1} \sigma_z^i \sigma_z^{i+1},$$

(4)

where $\omega_0$ is the energy difference ($\hbar = 1$) between the excited and ground states of an isolated spin (qubit), $J$ is the interaction constant, $\omega_1 \ll J \ll \omega_0$ is the amplitude of irradiation, and $\sigma_z$ and $\sigma^+$ are the Pauli operators. Qualitatively, the principle of operation of the model with the Hamiltonian (4) can be explained as follows. The weak resonant field with amplitude $\omega_1$ can flip a spin only if its two neighbours are in different states and the shifts of the resonance frequency, caused by interaction with these neighbours, are compensated. Therefore, if the initial state is all spins up, nothing happens. If the first spin is flipped, its neighbour becomes resonant and flips, then the next neighbour, and so on, generating the polarization wave. Of course, real multi-spin dynamics is more complex and all spins move simultaneously. In the limit $\omega_1 \rightarrow 0$ an analytical solution [5] is available for this model. Its results support the qualitative picture described above.

It is difficult to find a real physical system with the Hamiltonian (4). Simulations for short spin chains with more realistic Hamiltonians [6] showed that, even in the absence of relaxation and decoherence, the possibility to launch a strong polarization wave and reach efficient amplification critically depends on parameters of the spin Hamiltonian. Therefore, experimental demonstration of this phenomenon would be encouraging.

In liquid-state NMR, it is possible to find spin systems with Hamiltonians resembling (4). Isotropic $J$-couplings between the nearest spins are much stronger than between the remote spins. Therefore, a linear chain of nuclear spins may be almost a chain with the nearest-neighbour interactions. Truncation of the isotropic $J$-coupling to a ZZ-term can result from large difference between the chemical shifts of the neighbour spins. An unavoidable complication is a need for multi-frequency irradiation to irradiate each spin at its own resonance frequency. For the present experiment, we have chosen a chain of four $^{13}$C nuclear spins of fully $^{13}$C-labelled sodium butyrate. The major differences between its spin Hamiltonian (under proton decoupling) and the Hamiltonian (4) are the following. All spins have different resonance frequencies. $J$-coupling constants between the nearest neighbours are not all equal. There are small couplings between the next- and next-next-nearest neighbours. The values of the coupling constants are $J_{12} = 51.3$ Hz, $J_{13} = 1.9$ Hz, $J_{14} = 3.3$ Hz, $J_{23} = 33.8$ Hz and $J_{34} = 34.2$ Hz.
Figure 1. Line (a): thermal equilibrium $^{13}$C NMR spectra for the spins 1 to 4; (b) pseudopure state with all spins up; (c) the spectra after the first spin has been flipped; (d) the spectra after the evolution time $\omega_1 t = 5.2$.

The thermal equilibrium $^{13}$C spectrum is shown in figure 1, line (a). Interaction with the nearest neighbours splits the spectra of spins 1 and 4 into doublets, and that of spin 3 into a triplet. Four peaks of spin 2 result from different couplings to spins 1 and 3. Fine structure of the peaks comes from interactions beyond the nearest neighbours.

The first step of our experiment is the initialization of the system in the state with all spins up. This pseudopure state [7, 8] has been prepared by using a partial saturation [9]. The saturation has been performed with a seven-frequency pulse, which irradiated all allowed single-quantum transitions, except for the transitions with frequencies close to the transitions from the ground state (all spins up) to the four states with one flipped spin. As a result, there were no transitions from the ground state, and its population has been ‘trapped’, while the populations of the other 15 states have been equalized. The saturating pulse was 75 ms long and had an amplitude ($\gamma B_1 / 2\pi$) of 19 Hz per harmonic. A linear-response spectrum of the pseudopure ground state is presented in figure 1(b). It contains four peaks, one per spin, corresponding to transitions to the states with one flipped spin. The quality of the state is supported by the fact that the multiplet structure of the peaks disappeared. It is interesting that the peaks close to those in figure 1(b) have been eliminated even though these transitions were not saturated directly.

The result of flipping spin 1 with selective Gaussian pulse is shown in figure 1(c). It is a linear-response spectrum for the state with spin 1 down and spins 2–4 up. One can see that,
Figure 2. Polarizations of individual spins after the first spin has been flipped (bottom), and without flipping the first spin (top). They have been measured by integrating the spectra for each of the spins and normalized with respect to the pseudopure ground state in figure 1(b).

compared to figure 1(b), the peak for spin 2 ‘shifted’ to the left. This shift can be viewed as resulting from a change of the sign of the exchange field created by spin 1 when spin 1 flipped. One can also notice a small shift for spin 4. It results from the fact that the next-strongest interaction for spin 4, after its interaction with spin 3, is the interaction with spin 1, instead of spin 2.

The evolution has been driven by a three-frequency pulse with incremented length 0–150 ms. The three frequencies of irradiation marked by asterisks in figure 1(a) are the resonance frequencies for the spins 2–4 in the states where the left neighbour spin is down and the right neighbour spin (for spins 2 and 3) is up. The amplitude of the evolution pulse was 7.5 Hz for each harmonic. Evolution of polarizations of individual spins is shown in the bottom of figure 2. Spin polarizations have been measured by integrating the spectra in the entire spectral range for each of the spins and comparison with the pseudopure ground state in figure 1(b). One can clearly see the wave of flipped spins in figure 2, when first the spin 2, then spin 3, and finally spin 4 flip. A linear-response spectrum after the evolution time $\omega_1t = 5.2$ in figure 1(d) shows that in this state all spins are down. The result of the same experiment without flipping spin 1 is shown in the top of figure 2. Ideally, we would expect no changes in this case. Slow growth of
Figure 3. Evolution of the total polarization, triggered by a flip of the first spin (●), and without flipping the first spin (○); the lines are the results of numerical simulation.

polarizations is due to spin-lattice relaxation and the nuclear Overhauser effect, resulting from the proton decoupling.

Evolution of the total polarization of the four-spin system, with and without flip of the first spin, is shown in figure 3. The lines are the results of computer simulation. Deviations between the experimental and simulated dynamics are due to the relaxation and the Overhauser effect. An efficiency of the amplification dynamics can be described by a coefficient of amplification [5] defined as the ratio of the maximum change of polarization in a dynamics, triggered by a flip of a spin, to a direct change in polarization by a flip of a single spin. In our experimental implementation the measured coefficient of amplification is about 3. Another parameter describing the efficiency of amplification is the contrast, introduced in [2] and defined as the ratio of the total polarization change to the initial polarization. Its maximum value 2 corresponds to a complete flip of the total polarization. The value of the contrast obtained in our experiment was about 1.7.

In conclusion, a stimulated wave of polarization, triggered by a flip of the end spin, has been experimentally observed in a linear chain of nuclear spins. This ‘quantum domino’ dynamics is an explicit realization of a mechanism of quantum amplification.

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