Adiabatic transfer of coherences in a cluster of coupled nuclear spins

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It is experimentally demonstrated that quantum coherences can be efficiently transferred using adiabatic energy-level crossing. In a cluster of six dipolar-coupled proton spins of benzene, oriented by a liquid-crystalline matrix, a single-quantum coherence between one pair of states has been adiabatically transferred to another pair of states, and the superposition survived even after ten successive energy-level crossings.

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The quantum adiabatic theorem [1] tells that, when external parameters change very slowly, a quantum system prepared in an eigenstate of the initial Hamiltonian evolves into one of the eigenstates of the final Hamiltonian if the rates of nonadiabatic transitions between different instantaneous eigenstates are negligible during the evolution [2]. Population of a quantum state can be transferred to another state by adiabatic energy-level crossing, as schematically shown in Fig. 1. It is a well-known phenomenon, extensively studied both theoretically and experimentally for various quantum systems. Adiabaticity, i.e. conservation of entropy, and linearity of quantum mechanics suggest that superpositions of states should not be destroyed by such adiabatic switches. As an example, if a system is prepared in the superposition state $2^{-1/2}(|0\rangle+|1\rangle)$ and then, the population of state $|1\rangle$ is adiabatically transferred into the population of state $|2\rangle$, we would expect that the final state of the system will be the superposition state $2^{-1/2}(|0\rangle+|2\rangle)$.

Adiabatic transfer of superposition states has been studied for small atomic systems [3], and recent development of quantum computing attracted some attention to using adiabatic evolution for implementing quantum logic gates [4]. In the present work, for a system of strongly coupled nuclear spins, we experimentally demonstrate that coherences are conserved during adiabatic changes. Such transfer of coherences is expected to be useful for exploring complex quantum systems or implementing quantum computing algorithms.

According to the quantum adiabatic theorem, the state gains some dynamical phase factor, which is not detectable since the state is an eigenstate at all times. A general quantum state is a superposition of eigenstates, where a relative phase between individual eigenfunctions can be measured. Interesting questions are whether the coherences can be transferred by adiabatic energy-level crossing and what happens to the relative phases. As it is shown below, when populations of the states are adiabatically transferred via multiple energy-level crossings, not only the coherence is transferred but the relative phase between eigenstates can be preserved. The relative phase between two quantum states is observed as a phase of nuclear magnetic resonance (NMR) signal in our experiment.

The experiment has been performed with a Varian Unity/Inova 500 MHz NMR spectrometer. The sample contained 6% of benzene (Aldrich) dissolved in liquid-crystalline solvent MLC-6815 (EMD Chemical). In this system, each benzene molecule contains six proton spins coupled by residual dipole-dipole interactions. All intermolecular spin-spin interactions are averaged out by fast molecular motions. The system is, therefore, an ensemble of noninteracting spin clusters. The averaged longitudinal (T_1) and transverse (T_2) relaxation times are 2.1 and 0.48 s, respectively. Under radio-frequency (rf) irradiation, the Hamiltonian of the proton spins is

$$H = -\omega_0 \sum_{k=1}^{6} S_{kZ} - 2\omega_1 \sum_{k=1}^{6} S_{kX} \cos \omega t + \sum_{k>j>0}^{6} b_{jk}\left(S_{kZ}S_{jZ} - \frac{1}{2}S_{kX}S_{jX} - \frac{1}{2}S_{kY}S_{jY}\right), \quad (1)$$

where ω_0 is the resonance frequency of the nuclear spins, ω_1 and ω are the amplitude and the frequency of the rf field, and b_{jk} are the dipole-dipole coupling constants [5,6]. In the rotating frame, the Hamiltonian transforms into

$$H_{rot} = -\Delta\omega_H \sum_{k=1}^{6} S_{kZ} - \omega_1 \sum_{k=1}^{6} S_{kX} + \sum_{k>j>0}^{6} b_{jk}\left(S_{kZ}S_{jZ} - \frac{1}{2}S_{kX}S_{jX} - \frac{1}{2}S_{kY}S_{jY}\right), \quad (2)$$

where $\Delta\omega_H=(\omega_0-\omega)$ [5,6]. The adiabatic process has been

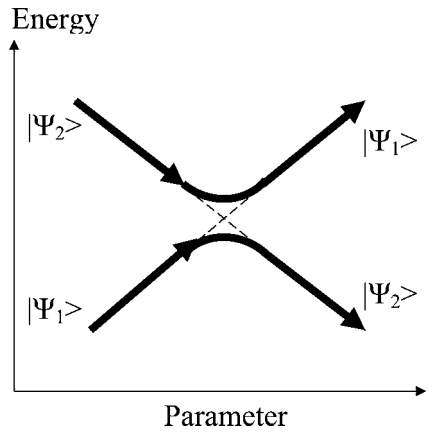

FIG. 1. Switching quantum states with adiabatic energy-level crossing.

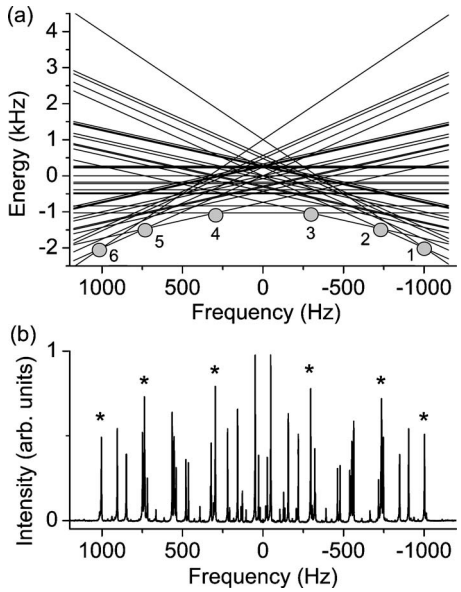

FIG. 2. (a) Energy diagram of benzene and (b) thermal equilibrium NMR spectrum.

implemented by slowly sweeping the frequency ω of the rf field. The amplitude ω_1 was sufficiently small to avoid undesired transitions between states.

Figure 2(a) shows the energy spectrum of a cluster of six dipolar-coupled proton spins of benzene as a function of the offset field $\Delta\omega_H$ in the rotating frame. Each eigenstate is characterized by its magnetic quantum number m, the z component of the total spin angular momentum, which determines the slope of each line in Fig. 2(a). Crossings of pairs of energy levels with $|\Delta m|=1$ correspond to allowed single-quantum transitions [7]. The thermal equilibrium nuclear magnetic resonance (NMR) spectrum displayed in Fig. 2(b) shows the frequency positions of allowed single-quantum transitions. The peak intensities are proportional to the transition probabilities. At thermal equilibrium, all differences of population between pairs of states with $|\Delta m|=1$ are equal in the high-field approximation ($|\omega_0| \gg |b_{jk}|$). The peaks marked by asterisks in Fig. 2(b) were identified as transitions between the states of the subspace of maximum total spin ($S=3$) by applying rf pulses to the pseudopure ground state with all spins up [8]. Strong rf pulse conserves the total spin of the system and when applied, as an example, to the pseudopure ground state, excites the transitions only in the $S=3$ subspace.

Suppose that the system is in one of its eigenstates and the rf field is turned on at some frequency far from any crossings. As the frequency changes, the system stays in the same state until it encounters an energy-level crossing. At the crossing, the rf field removes degeneracy and "switches" the populations of the levels as shown in Fig. 1. If the frequency changes further, one finds that the system stays in the second state. In this way, rf field with sweeping frequency causes a sequence of transitions from one state to another. As an example, suppose that the system is in the ground state and the rf field is turned on at the frequency -1100 Hz [Fig. 2(a)]. If the frequency increases adiabatically, the system stays in the ground state ($m=3$) until it encounters the crossing 1 in Fig. 2(a) where it switches to the other state, the first excited state with $m=2$. After that, it stays in this state until it meets the crossing 2 where it switches to the third state with $m=1$, and so on.

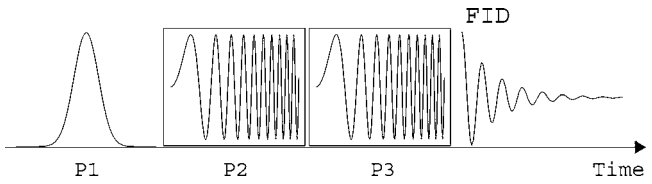

FIG. 3. Pulse sequence. 90° Gaussian pulse P1 of 50 ms duration excites the initial coherence between the ground ($m=3$) and the first excited ($m=2$) states. Adiabatic pulses P2 and P3 with linear frequency sweeping range of 2200 Hz and 40 ms duration transfer population of the state with $m=2$ to the state with $m=-3$ and population of the state with $m=3$ to the state with $m=-2$, respectively.

Figure 3 shows the pulse sequence used in the experiment. A single 90° Gaussian pulse P1 excites the initial coherence between the ground ($m=3$) and the first excited ($m=2$) states. The duration of this pulse was set to 50 ms to achieve selectivity. This single quantum coherence (superposition of $m=3$ and $m=2$ states) is directly observable. To discriminate between decoherence and nonadiabatic losses we introduced 80 ms delay, which is equal to the total duration of the two adiabatic pulses used to transfer the coherence. As a result of decoherence, the signal intensity after 80 ms delay decreased to 46% of its initial value. The signal is shown in Fig. 4(a). The pulse P2 (Fig. 3) with adiabatic frequency sweep transfers population of the $m=2$ state to the state $m=-3$ following the path 2-3-4-5-6 in Fig. 2(a). As a result, a superposition of the state $m=3$, which has not been affected by the pulse P2, and the state $m=-3$ is created. This

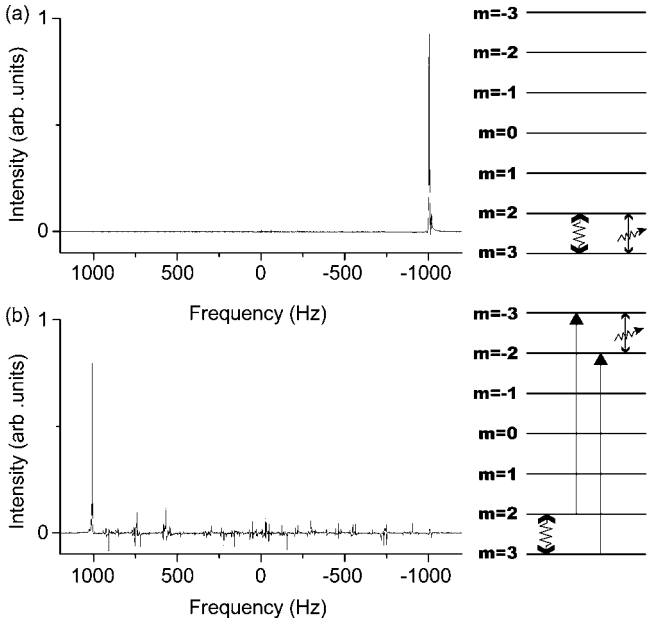

FIG. 4. Experimental results: (a) Single-quantum coherence excited by a Gaussian pulse; (b) coherence transferred by two adiabatic demagnetization pulses via ten energy-level crossings.

six-quantum coherence does not produce any NMR signal and cannot be observed directly. We applied the pulse sequence used in Ref. [8] and independently verified that the six quantum coherence was excited. This pulse sequence efficiently converts the six-quantum coherence into a mixture of two diagonal states with all spins up and all spins down, identified with a small-angle reading pulse. In addition, the transformation properties of the multiple-quantum coherence under rotations around z axis have been checked.

The pulse P3 (Fig. 3) transfers population of the $m=3$ state to the state $m=-2$ following the path 1-2-3-4-5 in Fig. 2(a). The resulting state, which contains a single-quantum coherence between the states $m=-2$ and $m=-3$, produces the NMR signal presented in Fig. 4(b). The intensity of the signal in Fig. 4(b) is 88% of that in Fig. 4(a).

Each of the two adiabatic pulses, P2 and P3, has linear sweeping range of 2200 Hz and 40 ms duration. They are shaped pulses with constant rf amplitude and 20 K steps of phase increment. The first of them starts at the frequency in the middle between the transitions 1 and 2 in Fig. 2(a), passes the frequencies of the transitions 2, 3, 4, 5, and 6, and ends at the frequency higher than that of the transition 6. The second adiabatic pulse starts at the frequency lower than the transition 1, passes the frequencies of the transitions 1, 2, 3, 4, and 5 and ends at the frequency midway between the transitions 5 and 6 in Fig. 2(a). The transitions caused by the adiabatic pulses are indicated by arrows on the simplified energy diagram in Fig. 4(b).

Recently, adiabatic evolution has been investigated as a potential method for quantum computation [9–11]. Adiabatic evolution may offer an efficient way of simulating other quantum systems with a quantum computer [12]. The adiabatic quantum computation has an inherent robustness against unitary control errors, decoherence and relaxation [12,13].

In conclusion, we have demonstrated that coherences can be successfully transferred by adiabatic evolution. In the system of six dipolar-coupled nuclear spins, quantum coherence has survived after ten consecutive adiabatic switches between states.

ACKNOWLEDGMENT

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