

Mechanical Multiplexer of Nuclear Spin States

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A spin $1/2$ is the simplest system that has been believed to support a single two-level quantum system, represented by a single resonance. We experimentally demonstrate that a spin- $1/2$ nucleus of ^{19}F in C_6F_6 exhibits an extra resonance corresponding to the emergence of the states, by mechanically rotating a sample and a coil in nuclear magnetic resonance (NMR) measurements. On the basis of the Floquet formalism, we identify the emergence of the extra two-level state due to the temporal periodicity generated by the mechanical rotation (mechanical spin multiplexing) and derive an operator algebra analogous to the planar rotor algebra in an effective description of the system. The observed multiplexing allows a single spin $1/2$ to carry more than two states and potentially enabling the processing of multiple quantum bits on a single spin.

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Introduction—The spins of nuclei and electrons have long been used as information carriers. Nuclear and electron spins are essential in quantum information technology, while magnetic memory devices have exploited electron-spin magnetization. A single spin $1/2$ is the simplest quantum two-level system, comprising only two states. Its energy absorption spectrum is thus of single resonance [1,2]; e.g., magnetic resonance spectra for spin- $1/2$ nuclei exhibit only a single peak as shown in Fig. 1(a), which corresponds to the transition from $|\uparrow\rangle$ to $|\downarrow\rangle$ states. There seems no chance for other resonance to appear in such a spin- $1/2$ system.

It is of fundamental importance to encode multiple pieces of quantum information in order to realize useful quantum information processing. Multiplexing of quantum states has opened up a new direction for such encoding by using only a single physical system. For instance, a single photon can carry information of a quantum bit (qubit) in several different forms such as polarization, path, temporal, and angular momentum degrees of freedoms [4–6]. Neutron interferometry has demonstrated the encoding of two and three qubits by utilizing the spin, path, and energy

of a neutron, thereby illustrating fundamental phenomena involving multientangled states [7–9]. Engineering such multiplexed states for other quantum systems remains one of the challenges in quantum information science.

In this Letter, we propose a method of multiplexing nuclear spin states combining a mechanical rotation and nuclear magnetic resonance (NMR). We show that, contrary to the common knowledge, a spin- $1/2$ nucleus of ^{19}F in C_6F_6 exhibits unknown extra resonance as shown in Fig. 1(b), when the NMR measurement is performed in the mechanical rotating frame (MRF) of reference same as the rotating sample under an external magnetic field non-colinear to the mechanical rotation axis. As shown in Fig. 1(b), the observed NMR line splits into three lines with the frequency of $\omega/2\pi$ and $(\omega \pm \Omega)/2\pi$, respectively. Here, ω and Ω represent the resonance frequency under the external field of B_0 and the mechanical rotation frequency of the sample, respectively. The line splitting is not due to the spinning side band commonly observed in the magic angle spinning (MAS) NMR, since the NMR frequency shift is much greater than the original NMR line width [10]. Therefore, the three NMR line splitting over the wide range of frequency shown in Fig. 1(b) cannot be explained by conventional approach. As we discuss in the following,

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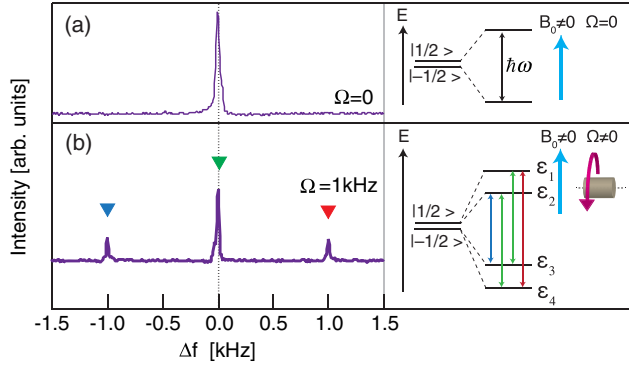


FIG. 1. NMR line splitting is created by mechanical rotation. (a) NMR line arises from $S = \pm 1/2$ state. (b) ϵ_{1-4} represent quasi-energy derived from Eq. (4) (see also Supplemental Material [3] Sec. 2.3). Origin of the horizontal axis is the resonance frequency without rotation, i.e., $\omega/2\pi = \gamma B_0$.

the extra resonance is clearly explained by the mechanical spin multiplexing (MSM).

Experimental method—We have developed a minute resonance circuit for NMR [11–14], to realize NMR measurements in a MRF of reference [Fig. 2(a)]. The sample is put into a coil (sample coil), which is used for picking up NMR signals from the sample. The sample coil is connected with another coil (coupling coil) and a chip capacitor in series to form a resonance circuit. The resonance circuit is installed into a high-speed rotor, which was commercially produced for MAS NMR measurements. The resonance circuit is mechanically rotated during the NMR measurements, in which the electromagnetic coupling is established between the coupling coil in the MRF of reference and the stationary coil in the laboratory frame of reference. The stationary coil is connected to an NMR spectrometer. A radio-frequency wave pulse generated at the NMR spectrometer is transmitted via the stationary and coupling coils to the sample coil. Then, an NMR signal picked up at the sample coil is transmitted via the coils to the spectrometer. We set the sample coil tilting from the rotation axis by the angle $\alpha \simeq 45^\circ$ to detect the longitudinal and transverse components of nuclear spin dynamics with respect to the mechanical rotation axis. To simplify the following discussion, we define the z axis as the mechanical rotation axis. The other coordinate axes of the laboratory and mechanical rotating frames of reference are defined as shown in Figs. 2(b)–2(d), respectively. In the laboratory frame of reference shown in Fig. 2(b), the external magnetic field is applied in the z - x plane, where θ represents the angle between the z axis and the direction of the magnetic field. The sample and the resonant circuit are rotated at the angular velocity Ω around the z direction. In the MRF of reference same as the rotating sample, the static external field is rotating with the frequency $-\Omega/2\pi$ and the sample is static as shown in Fig. 2(c). In addition, the Barnett field B_Ω emerges as an inertial field along the

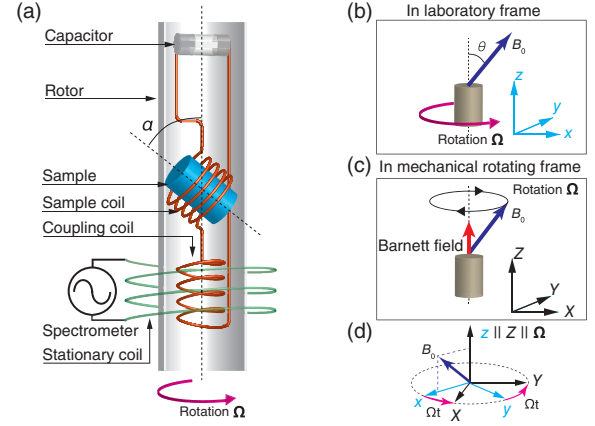


FIG. 2. A schematic illustration of the tuning circuit used in the present study. (a) The sample coil, the coupling coil, and the capacitor comprise a resonant circuit put inside the rotor. The sample coil tilts from the mechanical rotation axis [z (Z) axis] and the angle, α , is intentionally set to be nearly 45° . (b),(c) Experimental conditions observed from the laboratory and the mechanical rotating frames of reference, respectively. The notations of the coordinate axes are defined as small and capital letters for the laboratory and mechanical rotating frames of reference, respectively. The mechanical rotation axis is in parallel to the z (Z) axis and the external magnetic field is applied in the z - x plane of the laboratory frame of reference. (d) Comparison of the coordinate axes between the laboratory and mechanical rotating frames of reference.

mechanical rotation axis associated with the coordinate transformation [14–17]. We used liquid F_6C_6 with a purity of 99.0% as a sample for the NMR measurement.

Experimental results—Figure 3(a) shows ^{19}F ($S = 1/2$) NMR spectra for F_6C_6 measured by rotating the sample and the coil system simultaneously. The rotation axis is set perpendicular to the external magnetic field. When the sample is not rotating ($\Omega = 0$), we observe usual one NMR line arising from the two-level system of the nuclear spin $S = 1/2$ system [18]. When the sample is rotating ($\Omega \neq 0$), on the other hand, the single NMR line observed at $\Omega = 0$ splits into three lines, despite $S = 1/2$. The frequency shifts Δf deviated from the value of $\gamma B_0/2\pi$, where γ is the gyromagnetic ratio of the spin, coincide with 0 and $\pm\Omega/2\pi$ kHz, respectively.

When we put the sample coil perpendicular to the rotation axis ($\alpha = 90^\circ$), only the transverse components of nuclear spin dynamics with respect to the rotation axis can be observed. The result is shown in Fig. 3(b). The single NMR line splits into two accompanied by the NMR shift of $\pm\Omega/2\pi$. Therefore, these two lines can be assigned to the transverse components of the nuclear spin dynamics. By contrary, the NMR line at $\Delta f = 0$ shown in Fig. 3(a) can be assigned to the longitudinal components of nuclear spin dynamics with respect to the rotation axis. It should be noted that usual spinning NMR measurements have only observed this longitudinal component, and, thus, there

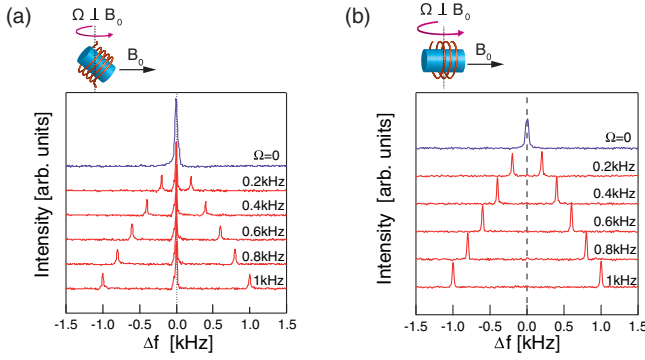


FIG. 3. ^{19}F NMR spectra in C_6F_6 obtained at various values of Ω with the external field perpendicular to the rotational axis. (a), (b) The NMR spectra obtained with the set up of the sample coil set 45° and 90° to the rotation axis, respectively. The horizontal axes Δf are the frequency measured from the center frequency of the NMR spectrum at $\Omega = 0$.

is no additional NMR shift accompanied by the sample rotation.

In Fig. 4(a), we show the θ dependence of the NMR spectra. The rotation frequency $\Omega/2\pi$ is fixed at $+1$ kHz. Contrary to the naive expectation based on the θ dependence of the Barnett field projected onto the external field direction, $B_\Omega \propto \cos \theta$, the observed resonance frequency does not depend on θ , but the NMR intensity does depend on θ : at $\theta = 0^\circ$, only the single NMR line is observed at $\Delta f = +1$ kHz. The intensity of the signal decreases with increasing θ from 0° to 180° . Around $\theta = 20^\circ$, the center

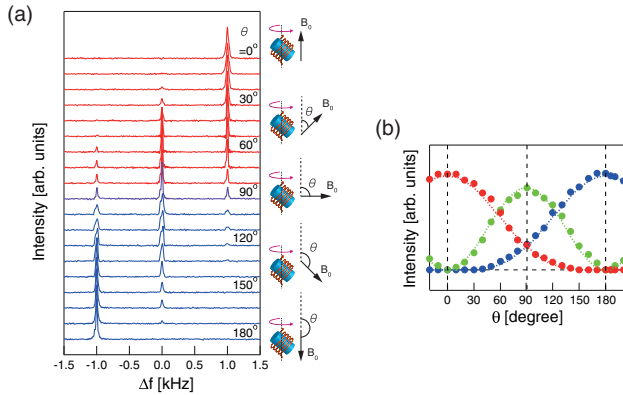


FIG. 4. Angular variation of the NMR spectra. (a) NMR spectra obtained at various values of θ , the angle between the mechanical rotation axis and the external field. The rotational frequency is fixed at $\Omega/2\pi = +1$ kHz. The horizontal axis Δf is the frequency measured from the center frequency of the NMR spectrum at $\Omega = 0$. (b) The angular dependence of the NMR intensities at the frequencies $\Delta f = 0$ (green circle), $\Delta f = +1$ kHz (red circle), and $\Delta f = -1$ kHz (blue circle), respectively. The dotted curves represent the NMR intensity obtained from the theoretical calculation [Eq. (5)]. From the fit to the data, the angle α between the mechanical rotation axis and the axis of the sample coil is estimated to be nearly 47° .

line appears at $\Delta f = 0$ kHz. The intensity of the center line shows maximum at $\theta = 90^\circ$. Then, around $\theta = 50^\circ$, the remaining signal appears at $\Delta f = -1$ kHz. The intensity of the remaining signal shows maximum at $\theta = 180^\circ$. The angle dependence of the NMR intensities are shown in Fig. 4(b).

The observed NMR spectra can well be explained by the transition between states related to the spin $S (= 1/2)$ and pseudospin T introduced to represent the rotation effect. The pseudospin T arises from the temporal periodicity of the quantum system introduced by the rotation. In the present study, the sample coil is in the MRF of reference. Therefore, the static external field is rotating in the MRF of reference. This rotating external field gives temporary periodic field for the nuclear spin system. In the following, we show that the effect of the periodically rotating field acting on the spin $1/2$ can be represented by a pseudospin T , whose transition can well reproduce the observed NMR spectra.

Theoretical analysis—We consider a spin $1/2$ exposed to an external magnetic field, $\mathbf{B}$, with the magnitude of B . The energy of the spin $\mathbf{S}$ is given by a formula called the spin Hamiltonian,

$$\hat{H}_{\text{spin}} = -\gamma \hbar \mathbf{B} \cdot \mathbf{S}. \quad (1)$$

The quantum eigenstates of Eq. (1) are up ($\uparrow$) and down ($\downarrow$) states. The spin system is then mechanically rotated perpendicular to the magnetic field with an angular velocity, Ω , to give temporal periodicity to the spin system, where γB is much greater than Ω . The spin is observed in the same MRF of reference. In the frame, the spin dynamics is described by time-dependent Hamiltonian, which has periodicity in time $\hat{H}_{\text{rot}}(t) = \hat{H}_{\text{rot}}(t + 2\pi/\Omega)$. The static quantum eigenstates cannot be obtained for the time-dependent Hamiltonian. However, for the time-periodic Hamiltonian, the steady states, which are eigenstates of the Floquet Hamiltonian [19–21], can be considered in the extended Hilbert space incorporating the degrees of freedom of Fourier modes originating from the periodicity. The orthonormal basis states in the extended Hilbert space are given by $|i, n\rangle = |\uparrow \text{ or } \downarrow\rangle \otimes |n\rangle$, where $|i\rangle = |\uparrow\rangle$ or $|\downarrow\rangle$ is the spin state, $|n\rangle = \sqrt{(\Omega/2\pi)} e^{in\Omega t}$ is the Fourier basis function, and n is an integer label of the Fourier modes. In the Floquet formalism based on the extended Hilbert space, the standard description of quantum mechanics with the static Hamiltonian, including spectral and perturbative analysis, is held over in the time-periodic case [19–21]. (See also Refs. [22–31] for recent applications of the Floquet formalism to solid-state, cold atomic, and photonic systems).

The effective spin Hamiltonian in the MRF of reference in the extended Hilbert space can be written as

$$\hat{H}_{\text{eff}} = \frac{\hbar \gamma B}{2} (\hat{L} + \hat{L}^\dagger) + \hbar \Omega (\hat{M} + \hat{N}), \quad (2)$$

where the spin-mode operators $\hat{L}$, $\hat{L}^\dagger$, $\hat{M}$ and the mode-number operator $\hat{N}$ are introduced. The operator $\hat{L}$ increases the spin and decreases the mode by single unit, $\hat{L}^\dagger$ decreases the spin and increases the mode by single unit, and $\hat{M}$ measures the spin of the states. The action of the spin-mode and mode-number operators on the basis states $|i, n\rangle$ in the extended Hilbert space is explicitly given in Supplemental Material [3] Sec. 2.3. The commutation relations among these operators become

$$\begin{aligned} [\hat{L}, \hat{L}^\dagger] &= 2\hat{M}, & [\hat{M}, \hat{L}] &= \hat{L}, & [\hat{M}, \hat{L}^\dagger] &= -\hat{L}^\dagger, \\ [\hat{N}, \hat{L}] &= -\hat{L}, & [\hat{N}, \hat{L}^\dagger] &= \hat{L}^\dagger, & [\hat{N}, \hat{M}] &= 0. \end{aligned} \quad (3)$$

This algebra has a similar structure to the planar rotor algebra (or orbital angular momentum algebra along a fixed axis), which is in contrast to the harmonic oscillator algebra. The cyclic property of the rotor has recently gained a renewed interest upon constructing bosonic quantum error correcting codes, since it is the underlying basis for recent developments of the variants of the Gottesman-Kitaev-Perskill codes [32–35].

Importantly, the operator $\hat{\Delta} = \hat{M} + \hat{N}$ commutes with $\hat{H}_{\text{eff}}$, and this leads to the conservation of the eigenvalue (Δ) of $\hat{\Delta}$ under the time evolution given by the effective Hamiltonian. Starting with the spin states in the static situation ($n = 0$), the effective Hamiltonian only allows the mixing between two pairs of the states: one is $|\mathcal{S}_1\rangle = |\uparrow, 0\rangle$ and $|\mathcal{S}_2\rangle = |\downarrow, 1\rangle$ with $\Delta = 1/2$, and the other is $|\mathcal{S}_3\rangle = |\downarrow, 0\rangle$ and $|\mathcal{S}_4\rangle = |\uparrow, -1\rangle$ with $\Delta = -1/2$. Thus, the extended Hilbert space can be reduced to the subspace spanned by these four states: $\mathcal{H}_{\text{sub}} = \text{Span}_{\mathbb{C}}\{|\mathcal{S}_i\rangle\}_{i=1,2,3,4}$. The reduced Hamiltonian in this subspace becomes

$$\hat{H}_{\text{red}} = \hbar\gamma B \left(\mathbb{I} \otimes \frac{\sigma_x}{2} \right) + \hbar\Omega \left(\frac{\sigma_z}{2} \otimes \mathbb{I} \right), \quad (4)$$

where $\mathbb{I}$ is the (2×2) unit matrix and σ_a ($a = x, y, z$) is the Pauli matrix. The form of $\hat{H}_{\text{red}}$ can also be expressed by the sum of two spin operators: one corresponds to the original spin represented by $\hat{S}_a = \mathbb{I} \otimes (\sigma_a/2)$, which gives the Zeeman coupling, and the other is the pseudospin $\hat{T}_a = (\sigma_a/2) \otimes \mathbb{I}$ which gives the rotational coupling. For general rotation with the angle θ between the mechanical rotation axis and external field, the spin states can also be comprised of the spin $\hat{S}_a$ and pseudospin $\hat{T}_a$. The eigenvalues (quasienergies) of the effective Hamiltonian in the extended Hilbert space are given by $\varepsilon_{1-4} = -(\hbar\gamma B \pm \hbar\Omega)/2$, and $(\hbar\gamma B \pm \hbar\Omega)/2$. Notably, the quasienergy spectrum is determined solely by the temporal periodicity of the Hamiltonian (and the external field) and is independent of the angle θ . Consequently, the resulting resonance frequencies exhibit no angular dependence, which is consistent

with the absence of angular variation in the shifts of the NMR spectra, as shown in Fig. 4(a).

From another perspective, we also show that the pseudospin generates from the topological origin based on the Berry's geometric phase [36,37]. Quantum states of a spin 1/2 with the mechanically rotating magnetic field has the Berry's gauge connection carrying a Dirac's monopole [38], and the representation of the gauge connection gives rise to a topological degree of freedom: one must choose either northern or southern hemisphere of the spherical surface swept by the spin [37,39]. The pseudospin degrees of freedom in the extended Hilbert space corresponds to the choice of the gauge connection. We present the detailed discussion about the topological nature of the pseudo spin in Supplemental Material [3] Sec. 2.5.

The NMR spectral intensity can be calculated by using Fermi's golden rule, which is generalized to the steady states in the extended Hilbert space, with a microwave perturbation given by $\hat{V}(t) = -\gamma\hbar e^{i\omega t} \mathbf{b} \cdot \mathbf{S} + (\text{H.c.})$. (See Supplemental Material [3] Sec. 3 for the details of the calculation.) The calculated power spectrum with the angle θ has three peaks at the frequency of $\gamma B/2\pi$, $(\gamma B \pm \Omega)/2\pi$, which are derived from the longitudinal and transverse components of nuclear spin dynamics with respect to the mechanical rotation axis, respectively. The proposed model explains well the experimental result shown in Fig. 3(a). The ratio of the peak power becomes

$$\mathcal{P}_{B+\Omega} : \mathcal{P}_B : \mathcal{P}_{B-\Omega} = \cos^4\left(\frac{\theta}{2}\right) : \sin^2\theta : \sin^4\left(\frac{\theta}{2}\right). \quad (5)$$

The angular dependence of the power spectrum well reproduces the experimental data shown in Fig. 4(b), demonstrating the relevance of the extra degree of freedom represented by the pseudospin T emerging in the present nuclear spin system.

The emergence of the extra resonance peak means the pseudospin state carries extra information in principle. Indeed, (logical) two qubits can be encoded in the multiplexed four states: $|\mathcal{S}_1\rangle = |0_L, 0_L\rangle$, $|\mathcal{S}_2\rangle = |1_L, 0_L\rangle$, $|\mathcal{S}_3\rangle = |0_L, 1_L\rangle$, and $|\mathcal{S}_4\rangle = |1_L, 1_L\rangle$. The independent unitary transformations on each qubit, $SU(2)_S \otimes SU(2)_T$, and the controlled-NOT gate, which leads to quantum entangled states [40], can be implemented in the two-qubit subspace $\mathcal{H}_{\text{sub}}$ by using the spin-mode and mode-number operators. Furthermore, these transformations can be physically realized by applying oscillating magnetic fields. (See Supplemental Material [3] Sec. 4 for details).

Summary—In this Letter, we have shown that a spin-1/2 nucleus of ^{19}F in C_6F_6 exhibits an extra resonance corresponding to the emergence of the multiplexed quantum states due to the mechanical rotation. These results indicate that one can run two-qubit quantum operations on a single spin 1/2 at a time, providing a new approach to quantum information science and technology. By using a more

elaborated combination of rotations, such as the double rotor around two rotation axes with different angular velocity [41], further multiplexing of spin states associated with the two rotation frequencies might be possible. Moreover, utilizing the similarity between the algebra of the spin-mode operators [Eq. (3)] and the planar rotor algebra, many qubits can be encoded in the infinite-dimensional extended Hilbert space [42]. Not only the mechanical rotation but similar multiplexing can also be expected when a spin is exposed to two effective magnetic fields with different directions and one of them is moving slowly. The observed effect will thus be a universal and powerful principle for breeding quantum states in condensed matter.

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