Controlled operations by solid-state NMR based on dipolar recoupling under magic angle spinning

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Abstract. A novel approach is proposed to perform controlled operations using solid-state NMR under magic-angle spinning (MAS), in which dipolar interactions are recovered by irradiating RF field or adjusting a spinning frequency. An important feature of this method is that the pulse sequences become simple, since the interactions between all spins are automatically decoupled by MAS unless intentionally recoupled. On the other hand, conventional implementations by liquid NMR requires continuous application of decoupling pulses, making the experiment considerably complicated. In this paper, pulse sequences are described to carry out controlled operations for heteronuclear as well as homonuclear spin pairs.

Keywords: solid-state NMR, dipolar interactions, magic angle spinning (MAS), recoupling

1 Introduction

Among several candidates for physical realization of quantum computing, nuclear magnetic resonance (NMR) has been the most successful, in which an ensemble of nuclear spins are used. It is conventional to demonstrate NMR quantum computing using liquid samples at room temperature, at which the spin system is, undesirably, in a highly mixed state. Although several ideas have been proposed to get around this fatal flaw by creating pseudo-pure states, the usage of the pseudo-pure states never promise efficiency gain over the classical computing. Thus, future progress in NMR quantum computing depends on whether the nuclear spin system can *physically* be initialized. So far, however, the prospect for realizing high nuclear spin polarization in the liquid state is beyond hope.

On the other hand, recent studies on dynamic nuclear polarization (DNP) using photo-excited triplet electron spins [1] have revealed that high nuclear spin polarization close to 1 can be attained in solid state organic materials even at moderate and convenient temperatures. In the hope of fully exploiting such a polarized solid system for NMR quantum computing, we propose a new approach to realize controlled operations in the solid state NMR. Our ideas, descrived in the following, is besed on the re-coupling of the decoupled dipolar interactions under magic angle spinning (MAS) of the sample, which was originally developed for structual analysis of molecules in chemistry.

2 A strategy for controlled operations in solid-state NMR

We propose the following steps to perform a Controlled-NOT (CNOT) operation in the solid-state NMR.

1. Erasure of the dipolar interaction between every pair of spins by magic angle spinning (MAS).
2. Selective recovery of the dipolar interaction.
3. Implementation of the CNOT gate using the recoupled dipolar interaction.

2.1 Erasure of dipolar interactions by MAS

Let us consider an isolated two spin-1/2 system I and S in the solid state. Under MAS, the dipolar interaction $\mathcal{H}_{IS}$ between these spins is represented as

$$\mathcal{H}_{IS} = D(t)\{I_Z S_Z - \frac{1}{4}(e^{-i\Delta t} I_+ S_- + e^{i\Delta t} I_- S_+)\}, \quad (1)$$

where Δ is the defference in their Larmor frequencies

$$\Delta = \omega_{\rm I} - \omega_{\rm S}, \quad (2)$$

and $D(t)$ is the spatial part of the interaction expressed as

$$D(t) = d[G_1 \cos(\phi + \omega_r t) + G_2 \cos(2\phi + 2\omega_r t)]. \quad (3)$$

G_1 and G_2 depend on the magnitude and orientation of the internuclear vector between the I and the S spins. d is the dipolar coupling constant and ϕ is the initial phase of the rotor.

The spin systems evolves under the action of the average Hamiltonian $\overline{\mathcal{H}_{IS}}$ defined by

$$\begin{aligned} \overline{\mathcal{H}_{IS}} &= \frac{\omega_r}{2\pi}\int_0^{2\pi/\omega_r} dt \mathcal{H}_{IS} \\ &= 0. \end{aligned} \quad (4)$$

Thus, the dipolar interaction does not have any first order effect on the time evolution of the system.

2.2 Recoupling of dipolar interactions under MAS

So far, many methods have been developed to recover the dipolar interactions under MAS. Here we deal with the two simplest methods, namely, *rotary* resonance recoupling [2] and *rotational* resonance recoupling [3]. The former is a technique to recouple heteronuclear dipolar interactions, and the latter is to recouple homonuclear dipolar interactions.

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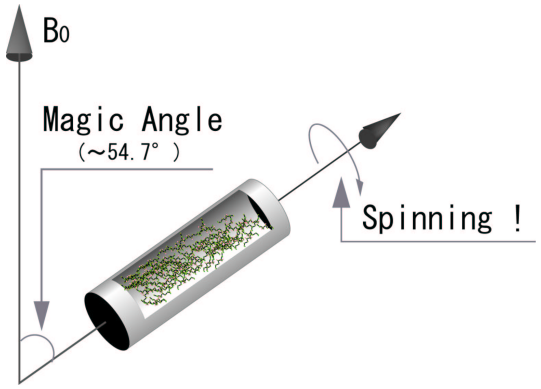

Figure 1: Magic angle spinning (MAS). A typical diameter of the rotor (sample container) is 5 mmϕ, and the sample can be rotated at a rate of up to ca. 18 kHz.

2.2.1 Rotary resonance recoupling (heteronuclear version)

In the case of rotary resonance for heteronuclear recoupling, an RF irradiation is applied at the Larmor frequency of the I spin with the intensity of ω_1. Then, I_z is modulated so that

$$I_z \to I_z \cos\omega_1 t + I_x \sin\omega_1 t. \tag{5}$$

When the intensity ω_1 of the irradiation field is adjusted to the spinning frequency ω_r, an interference occurs between the spatial part and the spin part in $\mathcal{H}_{IS}$, yielding a non-vanishing average Hamiltonian written as

$$\overline{\mathcal{H}_{IS}} = \frac{d}{2} G_1 I_z S_z. \tag{6}$$

Thus, the $I-S$ heteronuclear dipolar interaction is recovered under MAS.

2.2.2 Rotational resonance recoupling (homonuclear version)

In the case of rotational resonance for homonuclear recoupling, the spinning frequency ω_r is adjusted to the difference Δ in the Larmor frequencies, i.e.,

$$\omega_r = \Delta. \tag{7}$$

Then, time-independent components arise in $\mathcal{H}_{IS}$, leading to a non-vanishing average Hamiltonian of the form

$$\overline{\mathcal{H}_{IS}} = -\frac{1}{8} d G_1 (I_+ S_- + I_- S_+). \tag{8}$$

2.3 CNOT operation

Since the recoupled heteronuclear dipolar interaction (Eq. (6)) has the same form as J couplings conventionally utilized in liquid NMR quantum computing experiments, a CNOT gate operation can be realized in the same way as in liquid experiments for the case of the heteronuclear dipolar recoupling. For the less familiar case of homonuclear spins (Eq. (8)), one can either follow prosedures proposed recently [4, 5], or construct the CNOT gate by oneself, as shown in Fig. 2 (b).

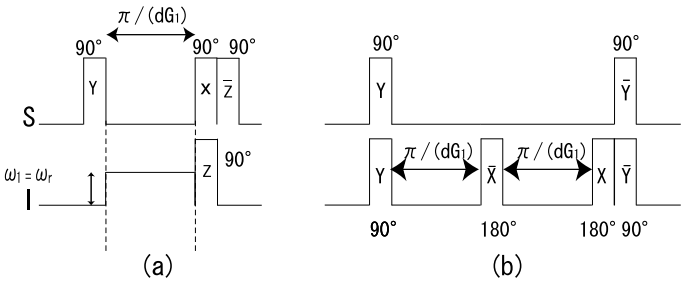

Figure 2: Examples of pulse sequences of the CNOT gate for (a) heteronulear and (b) homonuclear cases.

3 Summary

In this work, a novel approach has been proposed to realize controlled operations in the solid state NMR. An important feature of this method is that pulse sequences become simple, since the interactions between all spins are automathcally decoupled by MAS unless intentionally recoupled. On the other hand, conventional implementations by liquid NMR requires continuous application of decoupling pulse, making the experiment complicated considerably. The recoupling experiments are under progress and will be presented elsewhere.

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