

Attainment of high nuclear spin polarization in molecular crystal and its applicability to NMR quantum computing

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Abstract. An experiment is introduced to attain high nuclear spin polarization in molecular crystal by means of dynamic nuclear polarization using electron spins in the photo-excited triplet state, and the applicability of this technique to NMR quantum computing with physically initialized states is discussed.

Keywords: NMR, dynamic nuclear polarization (DNP), photo-excited triplet electron spins

1 Introduction

All demonstrations of NMR quantum computing reported so far rely on the creation of pseudo pure states from highly mixed states under thermal equilibrium [1, 2, 3, 4, 5, 6, 7], at the cost of exponentially decreasing detection sensitivity with increasing the number of qubits. Moreover, such NMR experiments are believed not to exhibit quantum behavior, since the density matrix with low spin polarization is separable [8]. Thus, in order for NMR to be more than a demonstration and survive hereafter as a means for quantum computing, it is essential to realize a *physically-initialized* state, or in other words, an initial state with very high nuclear spin polarization.

In this respect, dynamic nuclear polarization (DNP) [9] attracts considerable interest, in which large electron-spin polarization is transferred to nuclear spins. For DNP experiments, samples are doped with paramagnetic impurities or guest molecules photo-excitable to the triplet state as the source of spin polarization. The latter, called the “triplet DNP” henceforth in this paper, has some advantages over DNP using paramagnetic impurities; it allows us to prepare high nuclear spin polarization even at moderate and convenient temperatures. Moreover, the quantum gate operations would not be disturbed by the electron paramagnetism, since the triplet state decays to the diamagnetic ground state in a short lifetime. In this work, we briefly describe the triplet-DNP experiments which has recently been studied by one of the authors (K.T)¹, and discuss its applicability towards NMR quantum computing.

2 Triplet DNP

Fig. 1 shows simplified electronic energy levels of a molecule photo-excitable to the triplet state via intersystem crossing (ISC). A laser pulse excites the electronic state of the molecule from the ground state S_0 to the excited singlet state S_1 . The electronic state of a part of the excited molecules is transferred to the lowest triplet state T_1 , where the spin-orbit coupling very selectively populates the triplet sublevels independently of temperature and magnetic field strength. Thus

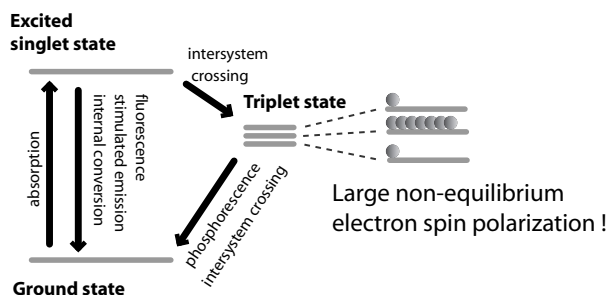


Figure 1: Electronic energy levels of a molecule photo-excitable to the triplet state.

non-equilibrium high electron-spin polarization can be prepared even at high temperatures in low magnetic fields, and transferred to ^1H spins by various DNP techniques such as the solid effect [10, 11], level anti-crossing [12, 13], or cross polarization [14, 15]. The most efficient DNP technique was proposed by Henstra et al. [15], in which an external magnetic field is adiabatically swept over the ESR line broadened by hyperfine couplings between the surrounding nuclear spins, during which microwave irradiation is applied. With an appropriate microwave intensity, the Hartmann-Hahn condition is fulfilled between the individual electron spin packets in the rotating frame and the ^1H spins in the laboratory frame one after another on the way to field-sweeping. This technique, which we call Integrated Cross Polarization (ICP) (originally called Integrated Solid Effect (ISE)), enables the very efficient polarization transfer, because it allows all the electron spin packets to constructively participate in the polarization transfer during the adiabatic sweep.

The ^1H polarization thus created is localized around triplet guest molecules first, and then it spreads over further host molecules by spin diffusion [16]. By repeating the ICP sequence, the ^1H polarization is successively pumped and stored along the static magnetic field. This triplet DNP technique has successfully been applied to polarize ^1H spins not only in a single crystal sample [17, 18], but also in a polycrystalline sample [19]. In the single crystal work, Takeda et al. realized the ^1H polarization of 0.7 in pentacene-doped naphthalene at 105 K in the magnetic field of 0.32 T [18]. The attained ^1H polarization of 0.7 reaches the maximum possible value given by the electron-spin polarization (~ 0.7) in the photo-excited

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¹The work on triplet DNP has been done at Department of Chemistry, Graduate School of Science in Kyoto University with T. Terao and K. Takegoshi [16].

triplet state of pentacene and is 2.1×10^5 times as large as the thermal equilibrium ^1H polarization. The successful buildup to the ultimate value is ascribed to a very careful consideration and optimization of the experimental conditions, such as the concentration of the triplet-molecules, the crystal orientation, the light-penetration depth, the width and duration of the field-sweep, the microwave intensity, and so on [16, 18].

3 Toward the application to NMR quantum computing

As mentioned above, two distinct processes contribute to the buildup of nuclear spin polarization in the triplet DNP experiment[16]; the direct polarization-transfer from the triplet-electron to the proton spins occurs within each triplet molecules, and the intermolecular polarization transfer is caused by spin diffusion among the spins on different molecules. This insight into the underlying processes behind the buildup behavior is helpful for strategizing experiments, so as to meet the needs for various possible applications, whose scope is not restricted to quantum computing.

Our idea towards the application to NMR quantum computing is to implant the molecules that are photo-excitabile to the triplet state in a magnetically inert single crystal matrix, and to make the ensemble of these triplet molecules act as a quantum computer. This idea is based on consideration of the following requirements.

- Material to be used must contain the triplet molecules. Moreover, in order to attain high-nuclear spin polarization by the triplet-DNP technique, it should be single crystal. Another reason for the necessity of single crystal is to make nuclear spins magnetically-equivalent for each atomic site on individual molecules, because in solid-state NMR, unlike in liquid NMR, anisotropic interactions are dominant.
- Nuclear spin-spin interactions between *different* molecules are not desirable, because they introduce unnecessary resonance line broadening and fast decoherence.
- The spin-spin interactions *within* each molecule must be present. In the presence of *intramolecular* interactions and in the absence of *intermolecular* interactions, the NMR spectrum would be well-resolved, and controlled operations can also be performed using dipolar interactions between nuclei within individual molecules.

To summarize, high nuclear spin polarization is a prerequisite for further progress in NMR computing, and the triplet DNP technique can be a solution to it. Since triplet DNP can only be applied to solid samples, it is necessary to develop a novel system that enables solid-state NMR quantum computing together with the triplet DNP experiments. We have proposed, as described above, one possible strategy towards the application of the triplet DNP technique to NMR quantum computing, which would enable us to perform true quantum computing using NMR.

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