Study of Electromagnetically Induced Transparency using long-lived Singlet States

Soumya Singha Roy and T. S. Mahesh*
NMR Research Center,
Indian Institute of Science Education and Research (IISER),
Pune 411008, India
(Dated: March 18, 2011)

The long-lived singlet states are useful to study a variety of interesting quantum phenomena. In this work we study electromagnetically induced transparency using a two-qubit system. The singlet state acts as a 'dark state' which does not absorb a probe radiation in the presence of a control radiation. Further we demonstrate that the simultaneous irradiation of probe and control radiations acts as a dynamical decoupling preserving the singlet state at higher correlation for longer durations.

Introduction

Simulating quantum phenomena on physical systems usually involves preparing a pure state, i.e., a definitely known quantum state and then carefully controlling its dynamics [1]. Nuclear Magnetic Resonance (NMR) is often regarded as a good candidate for studying quantum control [2]. Initializing an ensemble of nuclear spin systems into a pure state is nontrivial. Usually an indirect initialization can be achieved and is known as a pseudopure state [3]. Controlling the nuclear spin dynamics can be achieved with high fidelity using specially designed radio frequency (RF) pulses. Nuclear spins usually retain quantum coherences for durations long enough to carry out complex unitary evolutions [4]. Recently it has been established that singlet states of nuclear spin pairs under certain conditions can have extraordinarily long lifetimes [5, 6]. Singlet life-times often exceed single spin coherence times by an order of magnitude or more [7] and in a particular case the coherence lasted up to half an hour [8]. It has been demonstrated recently that such long-lived nuclear spin states can be utilized for the preparation of pseudopure states as well as to prepare high quality Bell states [9].

Electromagnetically Induced Transperency (EIT) involves rendering transparent a small frequency range of an absorption line of a quantum system due to the distrtuctive interference of transition probabilities induced by two electromagnetic fields [10, 11]. The importance of EIT stems from the fact that it is a purely quantum mechanical phenomenon and has no classical analogue. Several applications of EIT arise from the greatly enhanced non-linear susceptibility in the spectral region of induced transparency of the medium and the associated steep dispersion. In the following we first describe initializing a 2-qubit NMR system into a singlet state and then describe their applications for studying EIT.

Initializing Singlet States

The Hamiltonian for an ensemble of spin-1/2 nuclear pairs of same isotope, in the RF interaction frame, can be expressed as

$$\mathcal{H}^{\text{eff}} = h \left[\frac{\Delta\nu}{2} I_z^1 - \frac{\Delta\nu}{2} I_z^2 + J I^1 \cdot I^2 + \nu_{12} I_x^{1,2} \right]. \quad (1)$$

Here the RF frequency is assumed to be at the mean of the two Larmor frequencies, and $\Delta\nu$, J and ν_{12} correspond respectively to the difference in Larmor frequencies (chemical shift difference), the scalar coupling constant and the RF amplitude (all in Hz). In the limiting case of $\Delta\nu \to 0$, the system is said to have magnetic equivalence, and the singlet state $|S_0\rangle = (|01\rangle - |10\rangle)/\sqrt{2}$, and the triplet states $|T_1\rangle = |00\rangle$, $|T_0\rangle = (|01\rangle + |10\rangle)/\sqrt{2}$, and $|T_{-1}\rangle = |11\rangle$ form an orthonormal eigenbasis of the internal Hamiltonian [4]

$$\mathcal{H}_{\text{eq}}^{\text{eff}} = hJI^1 \cdot I^2. \quad (2)$$

The state $|S_0\rangle\langle S_0| - |T_0\rangle\langle T_0|$ can be easily prepared from the equilibrium density matrix $I_z^1 + I_z^2$ by using the pulse sequence shown in Figure 1. The initial state is of the form $|00\rangle\langle 00| - |11\rangle\langle 11|$. Let U be the propagator for the pulse sequence. It can be easily seen that $U|00\rangle = |S_0\rangle$ and $U|11\rangle = |T_0\rangle$ up to overall phases. Thus the pulse sequence in Figure 1 prepares a non-coherent statistical mixture of states $|S_0\rangle$ and $|T_0\rangle$.

The equivalence Hamiltonian may be realized by using a spin-lock [6, 12]. Levitt and co-workers demonstrated that under $\mathcal{H}_{\text{eq}}^{\text{eff}}$ the decay constant of singlet state $|S_0\rangle$ is much larger than T_1, and hence called it a long-lived state [5, 6]. Detailed theoretical analysis of singlet state decay have been provided by Levitt and co-workers [13, 14] and by Karthik and Bodenhausen [15]. Recently we had reported that high-fidelity singlet states can be easily prepared and characterized in two-spin systems under various experimental conditions [16].

*Electronic address: mahesh.ts@iiserpune.ac.in

FIG. 1: Pulse sequence for preparing singlet state

Due to the long lifetime of the singlet state, only $|S_0\rangle$ state survives after a sufficiently long spin-lock as illustrated in Figure 1 [16]. Based on this phenomenon, we had proposed that the singlet state can be used to efficiently initialize NMR quantum registers [9]. The pseudopure singlet state thus prepared can be used for further experiments on quantum information. In the following we describe the experimental demonstration of EIT using singlet states the dark state.

Electromagnetically Induced Transparency

EIT involves three states of a quantum system coupled by two electromagnetic radiations. If the system $(|01\rangle - |10\rangle)/\sqrt{2}$, then it is a stationary state under the two radiations and no net absorption or emission takes place [10, 11]. One of the two fields is known as 'control' and the other as 'probe' (see Figure 2). The stationary coherent state of the system under these two radiations is known as the 'dark state'.

EIT has been studied extensively in optical systems and has several applications such as enhancement of nonlinear optical frequency conversion [10], resonant four wave mixing [17], and coherent preparation of a maximal coherence [18]. More recently, exciting applications in quantum information processing have also been demonstrated [19, 20]. The group velocity of photons carrying quantum information can be made vanishing inside an EIT medium [21].

EIT has also been studied using NMR. Phase coherence of EIT has been studied by Murali et al using spin $7/2$ nuclei of ^{133}Cs system partially oriented in a liquid crystalline medium [22]. Cory and co-workers used a liquid state NMR system to demonstrated EIT [23]. These works involved an initial preparation of a longitudinal pseudopure state which was then converted into a dark state using a sequence of RF pulses.

In our demonstration using the pulse sequence in Figure 1, the dark state is naturally prepared because of the long-lived nature of the singlet state. After obtaining the maximum fidelity of the dark state, the equivalence Hamiltonian (spin-lock) is turned off and the probe and the control fields are applied simultaneously as shown in

FIG. 2: The energy level diagram indicates the three states of the 2-qubit system coupled by two electromagnetic radiations. The upper trace is spectrum obtained by applying only the probe field on the equilibrium state. The middle trace is that obtained by applying the probe and the control fields simultaneously. The bottom trace is the reference spectrum for the compound. Molecular structure of 5-chlorothiophene-2-carbonitrile is shown in the inset.

Figure 2. Further, in our implementation we have carried out the density matrix tomography of the dark states which enables us to quantitatively monitor the dynamics of the dark state.

Three levels $\{|00\rangle, |01\rangle, |10\rangle\}$ of the two ^{1}H spins of 5-chlorothiophene-2-carbonitrile were used to study EIT phenomenon (Figure 2). About 5 mg of the sample was dissolved in 0.75 ml of dimethyl sulphoxide-D6 and all the experiments are carried out in a Bruker 500 MHz NMR spectrometer at 300 K. The chemical shifts difference between these spins are 270.3 Hz and the scalar coupling is 4.1 Hz. The spin lattice relaxation time constant for the two spins obtained from inversion recovery experiment are about 6.3 s. RF spin-lock was achieved by WALTZ-16 modulations at 2 kHz amplitude. The lifetime of the singlet state was about 12.0 s, about two times the T_1 values of the individual spins implying the long-lived nature of the singlet state. The quality of the singlet state was estimated by the density matrix tomography [16]. The spin-lock duration of 15 s provided a correlation of about 0.991 with the singlet state.

As shown in Figure 2, the field on $|00\rangle \leftrightarrow |01\rangle$ transition is labeled as probe and that on $|00\rangle \leftrightarrow |10\rangle$ transition is labeled as the control. To study the effect of only the probe field, a simple Gaussian pulse of duration 520 ms was sufficient. The excitation spectrum of the Gaussian pulse is shown in the upper trace of Figure 2. The simultaneous irradiation of the probe and the control fields were achieved by a strongly modulated pulse of duration 240 ms, which was designed to be robust against the spa-
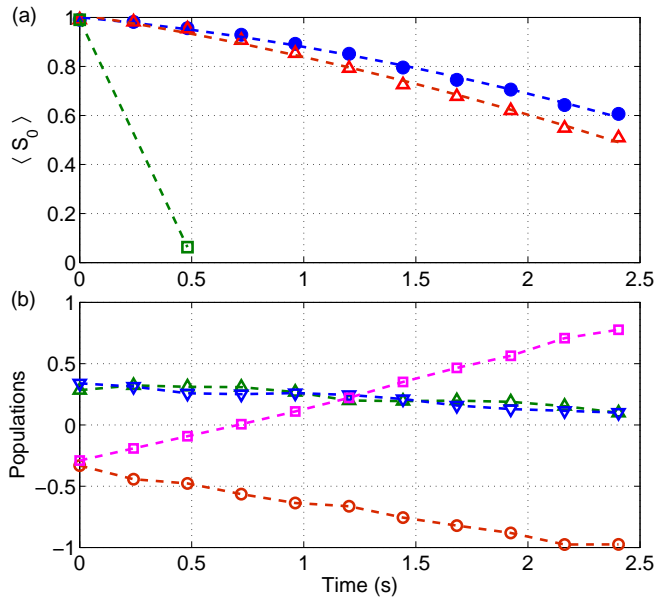
FIG. 3: (a) The correlation of the experimental dark state state under (i) no irradiation (triangles), (ii) under only the probe field (squares), and (iii) under the combined irradiation of probe and control pulses (filled circles). (b) The deviation populations of $|00\rangle$ (circles), $|01\rangle$ (down-triangle), $|10\rangle$ (up-triangle), and $|11\rangle$ (squares). The sum of all the deviation populations is zero as expected for a deviation density matrix.

FIG. 4: The correlation of the experimental dark state state under under the simultaneous irradiation of probe and control fields as a function of (a) offset and (b) relative amplitude of the probe irradiation. In (b) ν and ν_0 are the probe and control amplitudes. The duration of RF was 240 ms in all the experiments.

tial inhomogeneity of RF amplitude [24, 25]. Repeated application of this pulse allowed us to monitor the dark state over extended durations of time. The excitation spectrum of the simultaneous irradiation of the probe and the control is shown in the middle trace of Figure 2. The reference spectrum is in the lower trace of Figure 2. Of course, the selectivity of the probe and the control are not perfect. There are small amplitudes for the transitions connecting the $|11\rangle$ level, which should ideally have been left isolated.

We have monitored the state of the system at regular intervals using density matrix tomography. Figure 3a shows the correlation

$$\langle S_0 \rangle = \frac{\mathrm{trace}(\rho_{\mathrm{exp}} \cdot \rho_{\mathrm{th}})}{\sqrt{\mathrm{trace}(\rho_{\mathrm{exp}}^2) \ \mathrm{trace}(\rho_{\mathrm{th}}^2)}}. \tag{3}$$

of the experimental dark state ρ_{exp} with the theoretical singlet state $\rho_{\mathrm{th}} = |S_0\rangle\langle S_0|$.

Under the application of only control field, the dark state immediately collapses and the correlation of the experimental density matrix drops immediately. The dynamics in this case is expected to be oscillatory, and we have not studied this situation with further tomography. Under the simultaneous application of the probe and control fields, the dark state is maintained for much longer time. Ideally the dark state is a stationary state under the combined irradiation. But the decay of the experi-

mental dark state is due to the non-ideality of the control fields (limited selectivity and RF inhomogeneity) and due to decoherence of the dark state.

Figure 3 also shows the decay of the dark state under no irradiation. In this case the decay is slightly faster than that of the combined irradiation. Thus the combined irradiation of the probe and control fields acts as a dynamical decoupling of the dark state from the environment, storing it for longer duration of time [26, 27].

The density matrix tomography allows us to monitor the populations at various levels during the simultaneous application of probe and control [16]. As shown in Figure 3b, the populations of $|01\rangle$ and $|10\rangle$ remain equal and almost constant for up to two seconds, thus demonstrating the phenomenon of coherent population trapping. However, there appears to be a slow exchange populations between $|00\rangle$ and $|11\rangle$. This exchange is induced by the imperfect selectivity of the probe and control pulses.

We have also studied the dependence of the dark state correlation on the resonance offset of probe and on relative amplitudes of probe and control. These results are displayed in Figures 4a and 4b. It can be seen that the correlation is very sensitive to the offset of probe (and control). Correlation drops to below 0.1 by increasing the offset even by 1 Hz as shown in Figure 4a. Similarly from Figure 4b, it can be seen that the correlation is maximum when the amplitudes of the probe and control fields are equal.

Conclusions

Due to the long-lived nature, nuclear spin-pair ensembles can be conveniently initialized to singlet states with high fidelity. Treating the singlet state as a dark state under the simultaneous application of probe and control fields, we demonstrated electromagnetically induced transparency, which is a purely quantum mechanical phenomenon. It is shown experimentally that the dark state is retained for fairly long durations up to about two seconds. Monitoring the populations of various levels demonstrated the coherent population trapping and also indicated the effect of decoherence and of non-selectivity of the control and probe fields. Finally we have studied the sensitivity of the dark state to the offset and the relative amplitude of the probe with respect to the control field.

[1] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information, Cambridge University Press* (2002).

[2] C. A. Ryan, C. Negrevergne, M. Laforest, E. Knill, and R. Laflamme Phys. Rev. A 78, 012328 (2008).

[3] D. Suter and T. S. Mahesh, J. Chem. Phys. **128**, 052206 (2008).

[4] M. H. Levitt, *Spin Dynamics*, J. Wiley and Sons Ltd., Chichester (2002).

[5] M. Carravetta, O. G. Johannessen, M. H. Levitt, Phys. Rev. Lett. **92**, 153003 (2004).

[6] M. Carravetta and M. H. Levitt, J. Am. Chem. Soc. **126**, 6228 (2004).

[7] R. Sarkar, P. R. Vasos, and G. Bodenhausen, J. Am. Chem. Soc. **129**, 328 (2007).

[8] G. Pileio, M. Carravetta, E. Hughes, and M. H. Levitt, J. Am. Chem. Soc. **130**, 12582 (2008).

[9] S. S. Roy and T. S. Mahesh Phys. Rev. A **82**, 052302 (2010).

[10] Harris, S. E., J. E. Field, and A. Imamoglu, Phys. Rev. Lett. **64**, 1107 (1990).

[11] Harris SE, Phys Today, **50** 36 (1997).

[12] R. Sarkar, P. Ahuja, D. Moskau, P. R. Vasos, G. Bodenhausen, ChemPhysChem **8** 2652 (2007).

[13] M. Carravetta and M. H. Levitt, J. Chem. Phys. **122**, 214505 (2005).

[14] G. Pileio and M. H. Levitt, J. Chem. Phys. 130 (2009) 214501.

[15] K. Gopalakrishnan and G. Bodenhausen, J. Magn. Reson. **182**, 254 (2006).

[16] S. S. Roy and T. S. Mahesh, J. Magn. Reson. **206**, 127 (2010).

[17] Hemmer, P. R., D. P. Katz, J. Donoghue, M. Cronin-Golomb, M. S. Shariar, and P. Kumar, Opt. Letts. **20**, 769 (1995).

[18] Jain, M., H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996).

[19] R. G. Beausoleil, W. J. Munro, D. A. Rodrigues, T. P. Spiller J. Mod. Opt. **51**, 2441 (2004).

[20] Filippo Caruso, *Storing Quantum Information via Atomic Dark Resonances*, Master Thesis (2005), University of Catania, LANL arXiv:1001.4660

[21] M. Fleischhauer and M. D. Lukin, Phys. Rev. Lett. 84, 5094 (2000).

[22] K.V. R.M. Murali, Hyung-Bin Son, M. Steffen, P. Judeinstein, and I. L. Chuang, Phys. Rev. Lett. **93**, 033601 (2004).

[23] L. E. Fernandes, J. T. Choy, D. R. Khanal, D. G. Cory, Concepts inMagnetic Resonance PartA, Vol. 30A(5) 236245 (2007)

[24] E. M. Fortunato, M. A. Pravia, N. Boulant, G. Teklemariam, T. F. Havel and D. G. Cory, J. Chem. Phys. **116**, 7599 (2002).

[25] T. S. Mahesh and Dieter Suter, Phys. Rev. A **74**, 062312 (2006).

[26] L. Viola, E. Knill, and S. Lloyd, Phys. Rev. Lett. **82**, 2417 (1999).

[27] S. S. Roy, T. S. Mahesh, and G. S. Agarwal, arXiv:1102.3560v2.