Implementation of universal control on a decoherence-free qubit

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Abstract. We demonstrate storage and manipulation of one qubit encoded into a decoherence-free subspace (DFS) of two nuclear spins using liquid state nuclear magnetic resonance techniques. The DFS is spanned by states that are unaffected by arbitrary collective phase noise. Encoding and decoding procedures reversibly map an arbitrary qubit state from a single data spin to the DFS and back. The implementation demonstrates the robustness of the DFS memory against engineered dephasing with arbitrary strength as well as a substantial increase in the amount of quantum information retained, relative to an un-encoded qubit, under both engineered and natural noise processes. In addition, a universal set of logical manipulations over the encoded qubit is also realized. Although intrinsic limitations prevent maintenance of full noise tolerance during quantum gates, we show how the use of dynamical control methods at the encoded level can ensure that computation is protected with finite distance. We demonstrate noise-tolerant control over a DFS qubit in the presence of engineered phase noise significantly stronger than observed from natural noise sources.
1. Introduction

The ability to effectively protect the coherence properties of a quantum information processing (QIP) device against the detrimental effects of environmental interactions is a prerequisite for realizing any potential gain of quantum computation and quantum information theory [1]. Approaches based on noiseless (or ‘decoherence-free’ [2]) coding offer a promising venue for meeting the challenge of noise-tolerant QIP. The theory of decoherence-free subspaces (DFSs) has been the focus of intensive development particularly by Zanardi, Lidar and coworkers [3]–[9]. Recently, the DFS idea has been incorporated within the more general approach based on noiseless subsystems (NSs) [10]–[14], which recover DFSs and their benefits as special instances.

The primary motivation behind ‘passive’ noise control strategies relying on either DFSs or NSs is to take advantage of specific symmetries occurring in the noise process to single out subspaces or subsystems of the physical information processor that are inaccessible to noise. Once information is appropriately encoded into such noiseless structures, robust storage is ensured without requiring further active correction—as long as the underlying symmetry dominates. These features, together with their stability against symmetry-perturbing errors [5, 6, 8] and the consequent potential for concatenation with quantum error-correcting codes [7], make noiseless codes natural candidates as robust quantum memories. To date, experimental implementations include studies of DF states in quantum optical systems [15], and one-bit quantum memories based on both a DFS of two trapped ions [16] and an NS of three nuclear spins [17].

Achieving robust quantum information storage represents only a first, though indispensable, step toward the goal of reliable QIP. An important advance in this direction came from the identification of universality schemes, which in principle enable DFSs (or NSs) to support universal encoded quantum computation in a way that remains fully protected against noise. Both existential [11, 18] and constructive results [19] have been established. While the latter are especially appealing for a class of proposed quantum computing architectures governed by Heisenberg exchange interactions [20], implementations of these schemes remain difficult due to the stringent symmetry and tunability requirements on the control Hamiltonians.

Here, we take a first experimental step towards encoded quantum computation by demonstrating universal control over a one-bit DF quantum register of two nuclear spins. A novel key ingredient we use to implement encoded quantum gates is the combination of robust control design with the use of dynamical decoupling methods [21]–[24] directly on encoded degrees of freedom. Our results suggest that this may serve as a useful strategy for practically coping with the constraints required for DF computation.

The paper is organized as follows. In section 2 we review the collective decoherence model that is relevant to the work, along with the prescriptions from the DFS theory for both protected storage and manipulation of quantum information in a two-qubit system. In section 3, we outline our proposed approach to noise-tolerant control of DFS-encoded qubits based on concatenating encoded decoupling methods with robust control design. The general

† Although the terms ‘noiseless’ and ‘decoherence free’ have become essentially interchangeable in the QIP context, our preference is to associate decoherence effects with the irreversible spreading of quantum correlations that may proceed in the absence of energy exchange between the system and the environment—by including under the more generic ‘quantum noise’ term the possible occurrence of both decoherence and dissipation. In NMR terminology, we use ‘decoherence’ to mean a pure $T_2$ process corresponding to unrecoverable loss of phase relations over the ensemble.
principles are developed starting from the physical NMR setting relevant to the experiment. Section 4 contains an account of the control techniques used in the experiment and the reliability measures adopted to quantify the accuracy of the implementation. In particular, a notion of gate entanglement fidelity, generalizing Schumacher’s definition to allow a desired unitary evolution on the quantum data, is proposed and related to other fidelity metrics relevant to QIP. The experimental results demonstrating protected storage and universal protected quantum logic are presented and discussed in sections 5 and 6, respectively.

2. Protecting quantum information against collective decoherence

2.1. Collective decoherence

For a system $S$ composed of $n$ qubits, a purely decohering, collective interaction arises when the qubits couple symmetrically to a single environment $E$ and no exchange of energy takes place between $S$ and $E$. Physically, this model accounts for relaxation due to fully correlated fluctuations of the energy levels of each qubit—a situation that is approached if the qubits are close enough relative to the correlation length of the environmental coupling and the latter commutes with the natural Hamiltonian. Although not always applicable, this decoherence model has a practical significance for QIP. In particular, collective dephasing was shown to play a major role in quantum devices based on trapped ions [16]. In NMR systems, dephasing caused by fully correlated fluctuations of the local magnetic field provides the dominant relaxation mechanism of quantum coherences between identical species in sufficiently small, rigid molecules [25].

If $H = H_S \otimes I_E + I_S \otimes H_E + H_{SE}$ represents the Hamiltonian for the joint system plus environment, collective phase damping corresponds to an interaction Hamiltonian of the form

$$H_{SE} = J_z \otimes B_z,$$

where the operator $B_z$ acts only on $E$, and $J_z$ measures (in units $\hbar/2$) the projection of the total spin angular momentum along the quantizing axis $\hat{z}$. Thus, $J_z = \sum_{j=1}^{n} \sigma_z^j$ in terms of the Pauli operator $\sigma_z^j$ acting on the $j$th qubit. Starting from an initial state $\rho_{in} = |\Psi_{in}\rangle \langle \Psi_{in}|$, the system alone evolves after a time $t$ into $\rho_{in} \mapsto \rho_{out} = S(\rho_{in})$, where $S$ is the super-operator associated with $H$ [1]. Under the assumption that $S$ and $E$ are initially uncorrelated, $\rho_{out}$ can be expressed as

$$\rho_{out} = \sum_a F_a \rho_{in} F_a^\dagger, \quad \sum_a F_a^\dagger F_a = I_S,$$

for a set of Kraus operators $\{F_a\}$ [26]. Because $[H_S, J_z] = 0$ for pure decoherence, the unitary contribution due to $H_S$ can be separated out as $F_a = E_a \exp(-iH_{St})$, leaving a set of error operators $\{E_a\}$, that still satisfy $\sum_a E_a^\dagger E_a = I_S$, and describe the non-unitary effects of the environment. If $t$ is sufficiently short (or, equivalently, the coupling is weak enough), the operators $E_a$ can be expressed as linear combinations of the basic error generator $J_z$ and the identity. For arbitrary collective decoherence, the possible errors that the coupling (1) can induce belong to the interaction algebra [10] $A_z$ generated by $J_z$, i.e. the algebra containing all the linear combinations of arbitrary powers of $J_z$ and $I_S$. By construction, $A_z$ is Abelian, expressing the fact that pure decoherence is energy conserving—thus diagonal in the computational basis.

We focus on $n = 2$ qubits, in which case every element in $A_z$ can be written as a linear combination of three operators, $I, J_z = \sigma_z^1 + \sigma_z^2, J_z^2$—equivalently, we replace the latter with $\sigma_z^1 \sigma_z^2 = J_z^2/2 - I$ ($I = I_S$ henceforth). If $\{|00\}, |01\}, |10\}, |11\}$ denotes the
computational basis, an equivalent choice as a basis for $A_z$ are the three orthogonal projectors on subspaces with definite $\hat{z}$-angular momentum, $\Pi_{+2} = |00\rangle\langle 00| = (\mathbb{I} + J_z + \sigma_z^1\sigma_z^2)/4$, $\Pi_0 = |01\rangle\langle 01| + |10\rangle\langle 10| = (\mathbb{I} - \sigma_z^1\sigma_z^2)/2$, $\Pi_{-2} = |11\rangle\langle 11| = (\mathbb{I} - J_z + \sigma_z^1\sigma_z^2)/4$. In addition to labelling the basis states, the $J_z$ quantum number also allows classification of the possible transitions between these levels, via the so-called coherence order [25]. For a quantum coherence between two states with $\hat{z}$-angular momentum $k, \ell$, corresponding to the off-diagonal density matrix element $|k\rangle\langle \ell|$ ($k, \ell$ being now measured in units $\hbar$), the coherence order $m_{k\ell} = |k - \ell|$. The behaviour of the zero-, one- and two-quantum coherence orders present in a two-spin system undergoing collective decoherence can be described by obtaining an explicit set of error operators $\{E_a\}$. This is done starting from a unitary representation $U_{SE}$ for the joint $SE$ evolution [26],

$$\begin{align*}
|00\rangle e \rightarrow |00\rangle e_0, \\
|01\rangle e \rightarrow |01\rangle e_1, \\
|10\rangle e \rightarrow |10\rangle e_1', \\
|11\rangle e \rightarrow |11\rangle e_2,
\end{align*}$$

where the $|e\rangle, |e_k\rangle$ are generally non-orthogonal environment states, and the collective nature of the interaction sets $\langle e_1|e_1'\rangle = 1$. The overlaps $\langle e_0|e_1\rangle = \langle e_1|e_2\rangle = e^{-\gamma}, \langle e_0|e_2\rangle = e^{-\gamma'}$ parametrize the decay rates for single and double quantum coherences, respectively. By letting $\gamma' = 4\gamma (\gamma \geq 0)$ [25], and by using an orthonormal basis $|\mu_a\rangle$ obtained from the $|e_a\rangle, a = 0, 1, 2$, a choice of Kraus operators is given by $E_a = \langle \mu_a|U_{SE}|e\rangle$, i.e.

$$\begin{align*}
E_0 &= \Pi_{+2} + e^{-\gamma}\Pi_0 + e^{-4\gamma}\Pi_{-2}, \\
E_1 &= \sqrt{1 - e^{-2\gamma}}\Pi_0 + e^{-\gamma}(1 + e^{-2\gamma})\sqrt{1 - e^{-2\gamma}}\Pi_{-2}, \\
E_2 &= (1 - e^{-2\gamma})\sqrt{1 + e^{-2\gamma}}\Pi_{-2}.
\end{align*}$$

An equivalent representation was derived for dephasing quantum operations engineered via gradient-diffusion techniques in NMR [27]. In the limit of arbitrarily strong (or ‘crusier’) interaction, $\gamma \rightarrow \infty$, equations (2) simplify to $E_0 = \Pi_{+2}, E_1 = \Pi_0, E_2 = \Pi_{-2}$, leading to the full suppression of single and double quantum coherences. However, regardless how strong, collective decoherence produces no decay of the zero-quantum subspace spanned by $\{|01\rangle, |10\rangle\}$. Zero-quantum coherences and their properties have been long appreciated in NMR, with important applications in both high-resolution spectroscopy in inhomogeneous magnetic fields and contrast enhancement in magnetic imaging [28, 29]. Within NMR QIP, zero-quantum coherences are revisited in view of their natural potential to encode protected quantum information.

2.2. Decoherence-free encodings

In the DFS approach, the first step is ensuring that the quantum data to be protected are encoded into a DFS. Mathematically, a DFS is a subspace of the system’s state space spanned by a set of degenerate eigenvectors of all the error generators appearing in $H_{SE}$. For global dephasing on $n$ qubits as in (1), let $\mathcal{H}^{(J_z)}$ be the eigenspace corresponding to the eigenvalue $J_z$ of $J_z$, $J_z = n, n - 2, \ldots, -n + 2, -n$. Then states in $\mathcal{H}^{(J_z)}$ remain invariant under the environmental coupling,

$$J_z|\psi_L\rangle = J_z|\psi_L\rangle, \quad \forall|\psi_L\rangle \in \mathcal{H}^{(J_z)},$$

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which also implies a degenerate action of each error operator on the subspace:

\[ E_a |\psi_L\rangle = f_a |\psi_L\rangle, \quad \forall |\psi_L\rangle \in \mathcal{H}^{(j_s)}, \forall E_a \in A_z, \]  

for some coefficients \( f_a = f_a(j_s) \) fulfilling \( \sum_a |f_a|^2 = 1 \). If the system is initialized in a state

\[ \rho_{in} = |\psi_L\rangle \langle \psi_L| \in \mathcal{H}^{(j_s)}, \]

then

\[ \rho_{out} = \sum_a E_a e^{-i H s t} |\psi_L\rangle \langle \psi_L| e^{i H s t} E_a^\dagger = \left( \sum_a |f_a|^2 \right) e^{-i H s t} |\psi_L\rangle \langle \psi_L| e^{i H s t} = e^{-i H s t} \rho_{in} e^{i H s t}, \]

i.e. the evolution remains unitary within each \( \mathcal{H}^{(j_s)} \). Thus, each \( \mathcal{H}^{(j_s)} \) is a DFS under collective decoherence. The amount of quantum information that a given DFS is able to protect is determined by its dimension \( n_{j_z} \)—which is simply the degeneracy of the corresponding \( j_z \)-eigenvalue. In particular, for \( n \) even, the largest DFS is supported by the zero-quantum subspace \( \mathcal{H}^{(0)} \), with \( n_0 = n!/(n/2)!^2 \).

For \( n = 2 \) spins, \( \mathcal{H}^{(0)} \) is doubly degenerate, hence it provides the smallest DFS capable of protecting one qubit against collective decoherence. The robustness of this two-spin zero-quantum subspace under \( A_z \) has been explicitly derived above. In terms of basis states, our DFS qubit is defined by the encoding

\[ c_0 |0_L\rangle + c_1 |1_L\rangle = c_0 |01\rangle + c_1 |10\rangle, \quad \mathcal{H}_L = \mathcal{H}^{(0)} = \text{span} \{ |0_L\rangle, |1_L\rangle \}, \]  

for arbitrary complex coefficients \( c_0, c_1 \). It is worth stressing that the existence of a DFS is tied, at the physical level, to the occurrence of symmetries in the noise process. The way the latter reflect into the state space of the system both determines the possibility of invariant states as in (3), (4) and the associated degeneracies. For any interaction which is diagonal in the computational basis, the underlying ‘axial’ symmetry ensures that the individual \( \sigma^z_j \) are conserved quantum numbers. However, it is only under the additional permutation symmetry characterizing collective interactions that the degenerate conserved quantum number \( J_z \) arises—signalling the presence of a protected structure. The emergence of degenerate degrees of freedom preserved under the noise remains the key ingredient for more general DFSs [3, 5, 19] and, in still more elaborated forms, NSs as well [10, 12, 14, 17]. Taking advantage of the existing symmetries translates into major gains toward achieving noise-protected QIP. For instance, the simple two-bit encoding (5) preserves a qubit against collective dephasing of arbitrary strength, to be contrasted with independent phase errors—where protection can be achieved only with finite distance using a quantum error-correcting code.

### 2.3. Decoherence-free manipulations

Once protected storage of quantum information is obtained, the next step is to ensure that universal quantum gates are implemented without ever leaving the DFS. Because the states spanning a DFS are characterized by precise symmetry properties, symmetries are likewise crucial in determining the control operations to be applied for effecting DF quantum logic. Clearly, the allowed gates must map DFS states to DFS states. However, as any physical gate takes a finite time to execute, invoking unitary manipulations that preserve the DFS at the conclusion of the gate is not sufficient. To guarantee that the system remains within the DFS during the entire gating time requires the stronger condition that gates are generated by Hamiltonians that themselves respect the symmetry. Thus, the general problem requires identifying a universal set of Hamiltonians which satisfy the correct symmetry constraints and involve at most two-body interactions [19].
In our case, because a single DFS qubit is involved, this universal set of control Hamiltonians is composed of two observables generating an encoded $u(2)$ Lie algebra, exponentiation then giving the whole group $U(2)$ of encoded one-qubit transformations. A necessary and sufficient condition for a Hamiltonian $A$ to preserve the zero-quantum DFS can be obtained by demanding that $\langle 00 | A(c_0 | 01) + c_1 | 10 \rangle = 0$, $\langle 11 | A(c_0 | 01) + c_1 | 10 \rangle = 0$ for arbitrary $c_0, c_1$. This leads to the following matrix form for $A$ with respect to the computational basis:

$$A = \begin{pmatrix} a_1 & 0 & 0 & c \\ 0 & a_2 & b & 0 \\ 0 & b^* & a_3 & 0 \\ c^* & 0 & 0 & a_4 \end{pmatrix}$$

(6)

for real coefficients $a_j, j = 1, \ldots, 4$, and possibly complex $b, c$. In particular, this includes all the Hermitian operators belonging to the so-called commutant of the error algebra [10, 19], $A'_c = \{ X : [X, J_z] = 0 \}$, which collects all operators commuting with the noise. Because every operator in $A'_c$ can be represented as a linear combination of the identity, the one-bit operators $\sigma_j^2$, and the two-bit couplings $\sigma_1^x \sigma_2^y$, $\sigma_1^z = \sigma_1^x \sigma_2^x + \sigma_1^y \sigma_2^y + \sigma_1^z \sigma_2^z$ (Heisenberg coupling), Hamiltonians in $A'_c$ have $c = 0$—implying that all the DFSs are in fact preserved. An additional constraint (so-called independence [19]) can be imposed on Hamiltonians in $A'_c$ by also requiring $a_1 = a_4 = 0$, in which case $A$ has zero entries outside the selected zero-quantum DFS.

With respect to the DF encoding (5), a choice of operators that act as independent, encoded $\sigma_z, \sigma_x$ on $\mathcal{H}_L$ is given by

$$\sigma_z^L = L \frac{1}{2} (\sigma_1^z - \sigma_2^z), \quad \sigma_x^L = L \frac{1}{2} (\sigma_1^x \sigma_2^y - \sigma_1^y \sigma_2^x),$$

(7)

where the notation $=L$ means equality upon restriction to $\mathcal{H}_L$ and $\sigma_y^L = L i \sigma_z^L, \sigma_x^L$. The independence property is useful to allow parallel encoded manipulations on different DFSs. However, when only a single DFS is in use, requiring independence or even preservation of all DFSs has no advantages, and allowing for the most general Hamiltonian as in (6) may in fact increase the options available for implementation. For instance, an alternative choice for encoded $z$ and $x$ observables is

$$\sigma_z^L = L - \sigma_z^2, \quad \sigma_x^L = L \sigma_z^2, \quad \sigma_y^L = L i \sigma_z^L, \sigma_x^L / 2.$$ 

(8)

In principle, based on standard universality results [1], it is possible to generate any encoded unitary transformation by appropriately alternating evolutions under two Hamiltonians with the correct symmetry. For instance, this is certainly true if one can turn on/off a pair of Hamiltonians in $A'_c$ such as, say, $\sigma_1^z$ and the exchange interaction $E_{12} = (\sigma_1^z \cdot \sigma_2^z + \mathbb{I})/2$—for $i [E_{12}, \sigma_z^L]/2$ gives an encoded $\sigma_y^L$ and then $-i [E_{12}, \sigma_y^L]/2$ gives $\sigma_z^L$ as in (7).

Once encoded single-qubit manipulations are available, then universal encoded computation over DFS qubits requires the additional ability of implementing a non-trivial encoded gate between two logical qubits. For instance, a controlled-rotation gate could be constructed from a logical phase coupling of the form $\sigma_1^L / \sigma_z^L /$, which is supported by the natural couplings of many NMR and NMR-like Hamiltonians. We focus here on the first step of this programme, i.e. to obtain reliable single-qubit DF manipulations compatible with the constraints that QIP implementations unavoidably face in terms of both the form and the tunability of the available control Hamiltonians.

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3. Controlling encoded quantum information with reduced error rate

3.1. Two-spin NMR QIP as a case study

The total system Hamiltonian we consider, \( H_S \), is the sum of a time-independent internal Hamiltonian, \( H_{\text{int}} \), and a time-dependent external Hamiltonian, \( H_{\text{ext}} \). The internal Hamiltonian, composed of spin–field and spin–spin interactions, is [22]

\[
H_{\text{int}} = \pi (\nu_1 \sigma_z^1 + \nu_2 \sigma_z^2 + J \sigma^1 \cdot \sigma^2 / 2), \tag{9}
\]

where \( \nu_1, \nu_2 \) and \( J \) are the chemical shifts and the coupling constant, respectively. The external Hamiltonian, describing the interaction between the spins and an applied RF field, has the form [22, 30]

\[
H_{\text{ext}} = \sum_{k=1,2} e^{-i(\omega_{RF} t + \phi) \sigma_z^k / 2} (\omega_{\sigma^k_x} / 2)e^{i(\omega_{RF} t + \phi) \sigma_z^k / 2}, \tag{10}
\]

the transmitter’s angular frequency \( \omega_{RF} \), the initial phase \( \phi \) and the power \( \omega \) being tunable over an appropriate parameter range.

The implementation of an arbitrary unitary gate is accomplished by modulating \( H_{\text{int}} \) via an external control sequence. While sequences can be optimized numerically [30], average Hamiltonian theory (AHT) [31] provides a systematic method for describing any unitary propagator resulting from the evolution under the time-varying Hamiltonian \( H_S = H_{\text{int}} + H_{\text{ext}} \) in terms of an effective Hamiltonian \( \overline{H} \) applied over the same time interval:

\[
U(T) = T \exp \left( -1 \int_0^T d\tau H_S(\tau) \right) = e^{-i\overline{H} T},
\]

where \( T \) is, as usual, the Dyson time-ordering symbol. In particular, AHT underlies the design of coherent refocusing and decoupling methods, which are able to effectively turn on/off selected contributions to the average propagator over some time interval. These methods have been recently revisited within the QIP context in [11, 23, 24]. We recall that the basic idea is to subject the system to a cyclic train of pulses \( P = \{ P_j \}_{j=1}^M, \Pi_{j=1}^M P_j = I \) which, in the simplest setting, are assumed to be infinitely short and equally spaced by \( \Delta t > 0 \). The net controlled evolution over the period \( T = M \Delta t \) can then be expressed as

\[
e^{-i\overline{H} T} = \prod_{k=0}^M e^{-iH_k \Delta t},
\]

where the ‘toggling-frame’ Hamiltonians \( H_k \) are determined as \( H_k = U_k^\dagger H_{\text{int}} U_k \), in terms of the composite pulses \( \hat{U}_k = \Pi_{j=1}^k P_j, k = 1, \ldots, M, U_0 = I \) [25]. In the limit of sufficiently rapid control, \( \overline{H} \) simply approaches [23, 25]

\[
\overline{H} = \frac{1}{M} \sum_{k=0}^M H_k = \frac{1}{M} \sum_{k=0}^M U_k^\dagger H_{\text{int}} U_k.
\]

By appropriately designing the pulse sequence \( P \), undesired contributions to \( \overline{H} \) can be effectively turned off. For instance, a train \( P_1 \) of equally spaced, simultaneous \( \pi_x \) pulses on both spins (\( \pi_x^1, \pi_x^2 \) pulses) averages out any phase evolution due to the \( \sigma_z^j \) terms in (9). Similarly, the \( \sigma_x^1 \sigma_x^2 + \sigma_y^1 \sigma_y^2 \) coupling can also be averaged to zero by a pulse sequence \( P_2 \) consisting of repeated, equally spaced \( \pi_x, \pi_y \) pulses.
3.2. Universal gates via encoded dynamical control

While AHT represents a powerful tool for designing logic gates over physical, \textit{un-encoded} degrees of freedom, a direct application on DF-encoded qubits does not automatically result in DF manipulations. Even though ensuring that $\mathcal{H}$ has the general form (6) (for instance, $\mathcal{H} \in \mathcal{A}'_z$) leaves the system in a DF state, there is no guarantee that the control path has remained within the DFS at all intermediate times—possibly re-introducing exposure to noise.

We begin by noting that $H_{\text{int}}$ can be rewritten as

$$H_{\text{int}} = \pi \left( \frac{\nu_1 + \nu_2}{2} J_z + \frac{J}{2} \sigma^1_z \sigma^2_z + (\nu_1 - \nu_2) \sigma^L_z + J \sigma^L_x \right), \quad (11)$$

in terms, for instance, of the encoded observables (7)—which makes it explicit that $H_{\text{int}} \in \mathcal{A}'_z$.

Since both $J_z$ and $\sigma^1_z \sigma^2_z$ are constant on the code subspace $\mathcal{H}_L$, they can be ignored and $H_{\text{int}}$ further simplifies to

$$H_{\text{int}} = L \pi (\Delta \nu \sigma^L_z + J \sigma^L_x), \quad \Delta \nu = \nu_1 - \nu_2. \quad (12)$$

Thus, the natural evolution implements a non-trivial logical operation within $\mathcal{H}_L$. The challenge is to extract the required controlled operations by remaining, ideally, always within the DFS.

The situation is simpler in the limit where, as above, control pulses are treated as instantaneous. Because $H_{\text{int}} \in \mathcal{A}'_z$, one can ensure that each toggling-frame Hamiltonian $H_k$ also remains in $\mathcal{A}'_z$ by choosing pulses such that either $[U_k, J_z] = 0$ or $\{U_k, J_z\} = 0$. The latter condition is satisfied, for instance, by the above-mentioned pulse sequence $P_2$, which thus implements a net encoded identity in this idealized scenario.

Of course, the duration of real-life pulses is necessarily finite, and one needs to pay additional care to what happens during the pulse length [32]. In principle, DF logical operations can still be effected if sufficient control over the parameters $\Delta \nu, J$ in (12) is available. The general idea is to concatenate AHT with the underlying DF encoding, i.e. to implement refocusing \textit{directly with encoded rotations}. Let us look at our DFS qubit (a more expanded account is given in [33]; see also [34] for related work), and imagine that encoded $\pi^L_x$ pulses are available as $\pi^L_{x,y} = \exp(-i \pi \sigma^L_{x,y}/2)$. Then a sequence of equally spaced encoded $\pi^L_x$ pulses (in this case a Carr–Purcell sequence [21]) can be used to refocus the encoded phase evolution and only leave the encoded $\sigma^L_x$ coupling active in (12). This can be thought of as a logical or encoded ‘spin echo’ [35]. A similar procedure holds for extracting the encoded $\sigma^L_z$ Hamiltonian if encoded $\pi^L_z$ pulses are employed instead. Thus, the same schemes that are effective at turning on/off unwanted terms in the physical qubit evolution are effective at turning on/off unwanted terms in the encoded qubit evolution, provided ordinary control pulses are replaced with encoded ones. More generally, a group-theoretical framework extending the un-encoded approach of [23] to encoded dynamical decoupling can be constructed [33]. For our system, this implies that the ability to apply a \textit{single} Hamiltonian with the correct symmetry (e.g. $\sigma^L_z$) suffices, in principle, for gaining universal control.

Unfortunately, such control is not directly available in practice, as the evolutions induced by the external RF Hamiltonian (10) do not resemble, in general, evolutions under logical Hamiltonians. The approach we take results from the following compromise: we \textit{mimic} the implementation of a fully encoded refocusing scheme by using available pulses whose \textit{propagator} (not Hamiltonian) equals the required encoded rotation; we then compensate for the residual exposure to noise by control design. If pulse durations are optimized, then the system will reside in the DFS for a dominant portion of the computational time. In addition, pulse design can add
robustness against noise [36], reducing its impact while the system resides outside the protected space. While in the limit of weak noise with arbitrarily long correlation times these techniques provide robustness, for realistic noise models actual improvements will depend heavily on the noise parameters. An explicit implementation will be reported.

As already noted, these ideas open the way for manipulating more than a single encoded qubit. If, for instance, two DFS qubits are supported by the zero-quantum subspaces of, say, two proton and two carbon spins, the overall internal Hamiltonian will be expressible, to high accuracy and for a wide class of spin–spin coupling distributions, in terms of both single-qubit encoded observables $\sigma^L_{x,1,2}$ and the two-qubit encoded interaction $\sigma^L_1 \sigma^L_2$. Thus, the ability to separately control each encoded qubit via encoded refocusing, combined with the presence of the logical phase coupling, implies the potential of effecting universal quantum logic with reduced error rate.

4. Experimental outline

Liquid state NMR QIP techniques have been extensively discussed in the literature [22], and only the salient points are recalled here. Because the system exists in highly mixed, separable states, NMR QIP relies on ‘pseudo-pure’ (p.p.) states whose traceless, or deviation, component is proportional to that of the corresponding pure state. The identity component of the density matrix is unobservable and is treated as a constant under the assumption of unital dynamics (i.e. dynamics that preserves the completely mixed state). In this case, the evolution of a p.p. state is equivalent to the corresponding pure-state evolution. Initialization of the two-spin system into an intended p.p. state was accomplished using gradient-pulse techniques as described in [22, 37]. Throughout the experimental implementation, all deviation components were explicitly verified by state tomography [38]. A fixed amount of identity component that optimizes the fidelity between the experimentally determined and a desired reference p.p. state, $|00\rangle\langle00|$, was added to each reconstructed deviation density matrix. For each experiment, we prepared one of the p.p. input states $\rho^\text{p.p.}_{\text{in}} = |\psi_{\text{in}}\rangle\langle\psi_{\text{in}}|$, with $|\psi_{\text{in}}\rangle\langle\psi_{\text{in}}|$ providing a complete set of one-bit density matrices so as to allow quantum process tomography reconstruction [17, 39].

Our physical system is an ensemble of dibromothiophene molecules (figure 1) in a solution of CDCl$_3$. Measured values for the relevant parameters are listed in the caption of figure 1. The experimental procedure begins with the data qubit 1 containing the state $|\psi_{\text{in}}\rangle$ to be protected, $|\psi_{\text{in}}\rangle = c_0|0\rangle + c_1|1\rangle$, and the ancilla qubit 2 initialized to $|0\rangle$. Encoding of the initial input state to the code space $H^L$ is accomplished by the unitary transformation

$$U_{\text{enc}}(c_0|0\rangle + c_1|1\rangle)_1|0\rangle_2 = c_0|0_L\rangle + c_1|1_L\rangle,$$

where $U_{\text{enc}}$ is a controlled $\sigma_x$ rotation on bit 2 conditioned on bit 1 having the state $|0\rangle$. Next, an encoded operation is performed on the system in the presence of noise. The information is retrieved by applying a decoding transformation $U_{\text{dec}} = U_{\text{enc}}^\dagger$, producing a general output state of the form

$$\rho_{\text{out}} = U_{\text{target}}|\psi_{\text{in}}\rangle\langle\psi_{\text{in}}| U_{\text{target}}^\dagger = U_{\text{target}}|\psi_{\text{in}}\rangle_1 (U_{\text{target}}^\dagger \otimes |0\rangle_2) |\psi_{\text{in}}\rangle_1, \quad (13)$$

for a target single-qubit unitary transformation $U_{\text{target}}$ on the data spin. $U_{\text{target}} = I$ corresponds to storage of the quantum data under either engineered collective dephasing or natural noise, while $U_{\text{target}}$ is a non-trivial desired rotation for demonstrating universal quantum logic. All experiments were carried out on a 400 MHz Bruker AVANCE spectrometer.
5.10

Figure 1. Molecular structure of dibromothiophene. The two proton qubits are indicated. All experiments were carried out in a magnetic field of $\sim 9.7$ T with one proton on resonance. The frequency shift of the second proton is $\nu_2 = 137.5$ Hz, while the $J$-coupling constant is $J = 5.7$ Hz. The longitudinal and transverse relaxation times are $T_1 \sim 7$ s and $T_2 \sim 3.5$ s, respectively.

4.1. Unitary and non-unitary control

The un-encoded gate operations involved in the encoding and decoding networks were mapped into ideal pulse sequences using standard methods [22]. Pulses were then implemented by modulating $H_{\text{int}}$ with external RF fields as mentioned in section 3.1 [22].

Non-unitary evolution, either for p.p. state preparation or for emulating collective decoherence, were implemented using pulsed magnetic field gradients. Magnetic field gradients take advantage of the spatial extent of the sample to induce an incoherent evolution. Applying a gradient $\nabla_z B = \partial B_z/\partial z$ along the axis of the static field causes a linear variation of the Larmor precession frequency given by the spatially dependent Hamiltonian

$$H_{\text{grad}} = \gamma z J_z \nabla_z B / 2,$$

$\gamma$ being the gyro-magnetic ratio of the given nuclear species. This causes each quantum coherence $\rho_{k\ell} (k \neq \ell)$ to be multiplied by a spatially dependent phase factor, $\exp(-i\gamma z m_{k\ell} \nabla B_z \delta / 2)$, where $m_{k\ell}$ is the coherence order (defined earlier) and $\delta$ the duration of the gradient pulse. In other words, each part of the sample experiences a different coherent phase error. Tracing over the spatial degrees of freedom, as is done in an ensemble measurement, causes this incoherent evolution to become irreversible when considering the spin degrees of freedom alone. While the effects of this evolution could be immediately reversed, random molecular diffusion causes an irreversible spatial displacement that increases with both time and the molecular diffusion coefficient. Applying an inverse gradient after a time delay $\Delta$ (diffusion time) thus results in an exponential
decay of non-zero coherences, \( \exp(-\Delta/\tau) \), with an effective noise strength given by [37]

\[
\frac{1}{\tau} = D(\gamma B m_k \delta)^2,
\]

\( D \) being the diffusion coefficient of the sample. Note the scaling of this decoherence rate with the square of the coherence order, as anticipated in the derivation of equations (2).

Using these gradient-diffusion techniques, variable strength noise can be obtained by either changing the gradient strength or the diffusion time. It should be noted that, in both the incoherent and decoherent case, the induced phase error is collective to an extremely good extent, deviations from a collective action being determined by the product of the gradient strength (approximately 60 Gauss cm\(^{-1}\)) and the spatial displacement between the two hydrogen spins (on the order of angstroms).

### 4.2. Reliability measures for control

As a reliability measure quantifying the accuracy of implementing a target unitary transformation \( U \) on a system \( S \) we invoke a variant of the entanglement fidelity \( F_e \) as introduced by Schumacher [40]. In Schumacher’s notation, let \( R \) be an auxiliary ‘reference’ system, and let the initial entangled state \( |\Psi^{RS}\rangle \) of the pair \( RS \) be subjected to the overall evolution \( \mathbb{I}^R \otimes \mathcal{E}^S \). Starting from \( \rho^{RS} = |\Psi^{RS}\rangle \langle \Psi^{RS}| \), this produces a final state \( \rho^{RS'} = (\mathbb{I}^R \otimes \mathcal{E}^S)(|\Psi^{RS}\rangle \langle \Psi^{RS}|) \).

Then the entanglement fidelity of the process \( \mathcal{E}^S \) relative to the initial state of \( S \) alone, \( F_e = \text{Tr}_R(|\Psi^{RS}\rangle \langle \Psi^{RS}|) \), is defined as

\[
F_e(\rho^S, \mathcal{E}^S) = \text{Tr}\{|\Psi^{RS}\rangle \langle \Psi^{RS}| \rho^{RS'}\} = \text{Tr}\{|\Psi^{RS}\rangle \langle \Psi^{RS}| (\mathbb{I}^R \otimes \mathcal{E}^S)(|\Psi^{RS}\rangle \langle \Psi^{RS}|)\}, \tag{14}
\]

i.e. \( F_e \) measures the fidelity between the input and output states of the joint system: \( F_e = \text{Tr}\{|\Psi^{RS}\rangle \langle \Psi^{RS}| \rho^{RS'}\} \). \( F_e \) can be expressed in terms of quantities intrinsic to the system alone once an operator-sum representation for \( \mathcal{E}^S \) is available. If \( \mathcal{E}^S(\rho^S) = \sum_{\mu} A_{\mu}^S \rho^S A_{\mu}^{S\dagger} \), Schumacher showed that [40]

\[
F_e(\rho^S, \mathcal{E}^S) = \sum_{\mu} |\text{Tr}\{\rho^S A_{\mu}^{S}\}|^2. \tag{15}
\]

Because \( F_e(\rho^S, \mathcal{E}^S) = 1 \) if and only if \( \rho^{RS'} = |\Psi^{RS}\rangle \langle \Psi^{RS}| \) [40], \( F_e \) naturally quantifies the preservation of quantum information—perfect preservation corresponding to implementing \( \mathcal{E}^S = \mathbb{I}^S \). In the presence of the target transformation \( U \equiv U^S \neq \mathbb{I}^S \), the appropriate measure should equal unity if and only if \( \rho^{RS'} = U^S |\Psi^{RS}\rangle \langle \Psi^{RS}| U^{S\dagger} \). Thus, (14) is generalized to a gate entanglement fidelity as follows:

\[
F_e(U^S \rho^S U^{S\dagger}, \mathcal{E}^S) = \text{Tr}\{U^S |\Psi^{RS}\rangle \langle \Psi^{RS}| U^{S\dagger} (\mathbb{I}^R \otimes \mathcal{E}^S)(|\Psi^{RS}\rangle \langle \Psi^{RS}|)\}.
\]

By using the above operator-sum representation for \( \mathcal{E}^S(\rho^S) \), one can derive the equivalent expression

\[
F_e(U^S \rho^S U^{S\dagger}, \mathcal{E}^S) = \text{Tr}\{|\Psi^{RS}\rangle \langle \Psi^{RS}| (\mathbb{I}^R \otimes \tilde{\mathcal{E}}^S)(|\Psi^{RS}\rangle \langle \Psi^{RS}|)\} = F_e(\rho^S, \tilde{\mathcal{E}}^S),
\]

where the modified dynamical map \( \tilde{\mathcal{E}}^S = U^{S\dagger} \mathcal{E}^S U^S \) is defined by the set of transformed Kraus operators \( \{U^{S\dagger} A_{\mu}^S\} \). Thus, a perfect implementation of the desired gate \( U^S \) corresponds to perfect preservation of quantum information under \( \tilde{\mathcal{E}}^S \); the meaning of this is simply that, in the ideal case, the intended effect would be \( \mathcal{E}^S(\rho^S) = U^S \rho^S U^{S\dagger} \), which is equivalent to ensuring \( \tilde{\mathcal{E}}^S(\rho^S) = \rho^S \). Similar to (15), we then have

\[
F_e(\rho^S, \tilde{\mathcal{E}}^S) = \sum_{\mu} |\text{Tr}\{\rho^S U^{S\dagger} A_{\mu}^S\}|^2. \tag{16}
\]
where as above $\rho^S = \text{Tr}_R |\Psi^{RS}\rangle\langle\Psi^{RS}|$ is the initial density matrix of the system alone. Taking as the standard reference state a maximally entangled purification $|\Psi^{RS}\rangle$ for which $\rho^S$ is the fully mixed state, i.e. $\rho^S = I^S/N$ for a $N$-dimensional state space, (16) finally becomes

$$F_e(\tilde{\mathcal{E}}^S) = \frac{1}{2} \left( F_{U|0\rangle} + F_{U|+\rangle} + F_{U|+i\rangle} - 1 \right), \quad (18)$$

This form makes it explicit that the gate fidelity defined in [30] is identical with the gate entanglement fidelity formally introduced here. We shall still refer to the quantity in (17) simply as entanglement fidelity $F_e$ in the following.

The above reliability measure can be related to experimentally available data, and the results take particularly simple expressions in the case of single-qubit transformations we are concerned with. Starting from the standard maximally entangled Bell state for the joint $RS$ system, where $S$ and $R$ are now two qubits, and assuming that the process $\mathcal{E}^S \equiv \mathcal{E}$ actually implementing $U$ is unital and trace preserving, one finds

$$F_e = \frac{1}{2} \left( F_{U|0\rangle} + F_{U|+\rangle} + F_{U|+i\rangle} - 1 \right), \quad (18)$$

where $F_{U|\psi_{\text{in}}\rangle} = \text{Tr} \{ U|\psi_{\text{in}}\rangle\langle\psi_{\text{in}}| U^\dagger \mathcal{E}(|\psi_{\text{in}}\rangle\langle\psi_{\text{in}}|) \}$ for a generic one-bit pure input $|\psi_{\text{in}}\rangle$, and $|0\rangle$, $|+\rangle$, $|+i\rangle$ are eigenstates with positive eigenvalue of $\sigma_z$, $\sigma_x$, $\sigma_y$, respectively. This expression was used in [17] for the special case $U = I$. As a further remark, it is worth noting that $F_e$ as given in (18) is related to the so-called average gate fidelity $\bar{F}$ proposed in [41] via $\bar{F} = 2/3 F_e + 1/3$. Equation (18) is directly applicable to quantifying the accuracy of DF unitary manipulations as given, upon decoding, by (13).

5. Demonstration of a decoherence-free qubit

The utility of a DFS to preserve quantum information (i.e. to implement the identity operation) is demonstrated under the action of different classes of both engineered and natural noise: a variable-strength engineered decoherent noise, a full-strength (crusher) engineered incoherent noise, and the natural ambient noise due to relaxation. A significant improvement in the entanglement fidelity for each class of noise is seen. In addition, because the measured entanglement fidelities remain above the threshold value 0.50 [42], all implementations guarantee, in principle, the ability to preserve entanglement over a wide range of noise strengths. Unlike the case of an NS, the entire state of the system (data plus ancilla) remains unchanged under the action of the noise. While this was experimentally confirmed to a good accuracy†, we report the preservation of quantum information between the desired input state and the measured output state of the data qubit alone.

5.1. Engineered noise

Gradient-diffusion techniques were used to implement variable-strength noise. In order to isolate the effects of the applied noise, the time delay between encoding and decoding was kept fixed. In the first half of this time delay collective phase noise was applied to the system. Unwanted evolution due to the internal Hamiltonian was refocused during the second half of the delay by a pair of $\pi$ pulses $\mathcal{P}_2$ given in section 3.1. The gradient strength was varied over the full dynamic

† For all experiments, the ancilla spin experienced deviations from the intended state in the same range than the ones experienced by the data spin.
Figure 2. Experimentally determined entanglement fidelity for the implementation of variable-strength collective dephasing. Both the behaviour of the DFS-encoded (squares) and the un-encoded (circles) data is shown. The independent axis (noise strength) was determined by fitting the un-encoded data to an exponential decay of the form $F_e = A \exp(-t_{ev}/\tau) + 0.5$, with $t_{ev} = \Delta + 2\delta = 37.765$ ms. The un-encoded data are only displayed for reference. The encoded data are fitted to a constant value $F_e = C$, yielding the best estimate $C = 0.97 \pm 0.01$. Systematic uncertainties (not included in the figure) are $\sim 0.02$.

range of the spectrometer (0 to 60 Gauss cm$^{-1}$) and the diffusion time $\Delta$ was set in such a way that a significant amount of information was lost when the un-encoded data spin was directly exposed to noise. This was obtained by running separate experiments with encoding/decoding sequences turned off.

The experimental data are collected in figure 2. For the encoded data, assuming no additional loss of information with increasing noise strengths is consistent with the experimental results. Fitting the data with a constant value yields a value of $0.97 \pm 0.01$. The deviation from unity is consistent with the observed $F_e$ value for the reference situation of no applied noise (i.e. the one implementing a net identity evolution between encoding and decoding). Therefore, these losses are caused by imperfections in the applied pulses as well as by natural noise processes whose action is not in the correctable error algebra $A_z$ (see below). For all but the smallest noise values, a substantially increased amount of quantum information is retained using the DFS memory rather than leaving the system un-protected.

To further confirm the robustness of the DFS memory against arbitrary noise strengths, an incoherent implementation of all possible collective phase errors was also realized to emulate noise in the strong dephasing limit. A single magnetic field gradient pulse with maximum strength was applied, causing spins on the fringe of the sample to evolve through more than 750 cycles and therefore acquire large phase errors. Again, the loss of information in the presence of this crusher noise is compatible with the measured loss due to just encoding and decoding (see table 1).

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Table 1. Experimental data for the implementation of full-strength collective dephasing. Input–output fidelities and entanglement fidelities corresponding to the application of the intended error model to both the DFS-encoded ($Q_z, df$) and the un-encoded data spin ($Q_z, un$) are listed, along with the values relative to the reference situation of zero applied noise between DFS encoding and decoding ($Q_0, df$). Crusher gradient fields with full strength $\sim 60$ Gauss cm$^{-1}$ were applied for a period $\delta = 745$ $\mu$s. The measured values for the un-encoded test data confirm the expectation that the applied noise process induces full phase damping on the data spin, with predicted $F_e = 0.50$. Systematic uncertainties are $\sim 0.02$ while statistical uncertainties are $\sim 2\%$, both due to errors in the tomographic density matrix reconstruction.

| Quantum process | $F_{|0\rangle}$ | $F_{|+\rangle}$ | $F_{|+i\rangle}$ | $F_e$ |
|----------------|----------------|----------------|----------------|------|
| $Q_z, un$      | 1.00           | 0.50           | 0.50           | 0.50 |
| $Q_0, df$      | 0.98           | 0.95           | 0.94           | 0.93 |
| $Q_z, df$      | 0.97           | 0.98           | 0.96           | 0.95 |

5.2. Natural noise

The behaviour of both the DFS-encoded and the un-encoded data under ambient noise was also probed in a separate series of experiments, with the goal of gaining qualitative insight on the relevance of fully correlated dephasing in the naturally occurring phase relaxation processes. In this case, the holding time between encoding and decoding was varied to allow for a variable exposure to noise. Because natural relaxation takes significant contributions from $T_1$ processes (that are both amplitude and phase damping) in addition to transverse $T_2$ relaxation, the unitality assumption invoked in deriving the expression (18) for the entanglement fidelity is no longer accurate. While $F_e$ could be still evaluated directly from (17) upon experimentally extracting a set of Kraus operators, a simpler coherence metric is appropriate if the actual amplitude decay is of no interest. Similar to [16], the amount of quantum coherence $C$ (phase information) that is retained in the course of the noisy evolution can be quantified by experimentally determining the average off-diagonal component present in the output density matrix. Thus, corresponding to the two experimentally prepared transverse p.p. states $|+\rangle, |i\rangle$ defined above, we calculate

$$C = \frac{1}{2} \left( \text{Tr}\{\sigma_x E(|+\rangle \langle +|)\} + \text{Tr}\{\sigma_y E(|i\rangle \langle i|)\} \right),$$

where the map $E$ now corresponds to the natural noisy dynamics.

The experimental data are presented in figure 3. Holding times ranging from a fraction of a second up to a time scale comparable to $T_2$ were explored. An appreciable decay of the DFS qubit is seen in this case, witnessing the presence of non-collective phase-damping processes in the ambient noise. In spite of this non-robust behaviour, the DFS is still able to retain quantum coherence much longer than the un-encoded state. This implies that a significant contribution to the overall phase relaxation is actually caused by fully correlated dephasing, consistent with the physical intuition based on the geometrical and chemical structure of the molecule [43].

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Figure 3. Experimental data for the phase information retained after exposure to the natural system noise. The average preservation of $\sigma_x$ and $\sigma_y$ was measured as a function of holding times from 0 to $\sim$3 s. Improvement over the un-encoded case is seen, confirming that collective phase errors are one of the dominating modes of natural noise for this system.

6. Demonstration of universal control over a decoherence-free qubit

As stated in section 2.3, evolution according to two non-commuting encoded Hamiltonians is required for universal control. In the implementation, we found it convenient to adopt a choice of encoded observables intermediate between (7), (8), i.e. $\sigma^L_z = -\sigma^2_z$, $\sigma^L_x = (\sigma^1_x \sigma^2_z + \sigma^1_y \sigma^2_y)/2$ henceforth. In terms of this choice, and using the actual implementation parameters (see figure 1), the internal Hamiltonian (12) is given by

$$H_{\text{int}} = L -137.5\pi\sigma^L_z + 5.7\pi\sigma^L_x. \quad (19)$$

6.1. Encoded $z$ and $x$ rotations

According to the above expression, the strong $\sigma^L_z$ Hamiltonian dominates the $\sigma^L_x$ contribution. Therefore, an encoded $z$ evolution can be implemented, to high accuracy, by simply waiting the appropriate amount of time. Because of the negative sign in (19), a positive $\sigma^L_z$ rotation of $\theta$ can be implemented by rotating by $2\pi - \theta$ about the negative logical $z$ axis. A representative $\pi/2$ encoded rotation, $\exp(-i\pi\sigma^L_z/4)$, was experimentally implemented with a fidelity of entanglement of 0.94 ± 0.03. Comparing this result with the accuracy of the identity operation (see table 1), we see no significant loss of information due to the $z$ gate.

Implementing an $x$ rotation is less straightforward. While a logical $x$ Hamiltonian is present in (19), it is quickly averaged out by the stronger logical $z$ term. With the encoded system expressed in this form, it is clear that we must average out the $\sigma^L_z$ term at a rate faster than its strength [25], using control operations that commute with $\sigma^L_z$. The external RF Hamiltonian provides this extra control parameter, at the expenses of forcing the system to leave the DFS—if
only for short periods of time. While continuous irradiation of the spins would suffice to preserve the $\sigma^L_x$ Hamiltonian by matching the so-called Hartman–Hahn condition [44, 45], as discussed in section 3.2 a train of $\pi^L$ pulses achieves the same goal [21, 35, 46], with the advantage that the system may be left in the DFS for significant portions of the control time. The trick is to note that, although the evolution induced by the applied RF field does not resemble in general a $\sigma^L_x$ Hamiltonian, for the special case of a hard $\pi$ pulse the resulting propagator is

$$U_{h}^{1,2} = \exp\left(-i\frac{\pi}{2}(\sigma^1_x + \sigma^2_x)\right) = \exp\left(-i\frac{\pi}{2}\sigma^1_x\right)\exp\left(-i\frac{\pi}{2}\sigma^2_x\right) = -\sigma^1_x\sigma^2_x,$$

which mimics a net $\sigma^L_x$ operation: the action of $U_{h}^{1,2}$ on the code subspace is identical to the action of $\sigma^L_x$ as a unitary operator (not as a Hamiltonian—note that $\sigma^1_x + \sigma^2_x$ does not clearly respect the form (6)).

Composite pulses [47], which provide an excellent balance between speed and robustness, were used to implement each hard $\pi$ pulse. Six-period pulses optimized to be robust against variations in both chemical shift (phase errors) and RF strength (control errors) [36] were used to emulate the required sequence of encoded $\pi^L$. Each of these hard pulses is $62.4 \mu s$ in duration and is followed by a delay of $630 \mu s$. Therefore, the system resides in the protected space for over 90% of the computational time. The phases of the $\pi^L$ pulses were alternated systematically as described by a WALTZ sequence [48], so as to minimize the impact of experimental errors. In particular, a 64-cycle sequence was used to achieve a $\pi/2$ encoded rotation, $\exp(-i\pi\sigma^L_x/4)$, with a fidelity of entanglement of $0.94 \pm 0.03$. Again, we see no significant loss of information due to the $x$ operation.

### 6.2. Composite encoded $y$ rotation under collective phase noise

To explicitly test the robustness of the available logical $x$ and $z$ manipulations, a composite encoded rotation by $\pi/2$ about $y$ was implemented in the presence of variable-strength collective phase noise, i.e. the sequence of encoded rotations

$$\exp\left(-i\frac{\pi}{4}\sigma^L_y\right)\exp\left(-i\frac{\pi}{4}\sigma^L_x\right)\exp\left(i\frac{\pi}{4}\sigma^L_z\right) = \exp\left(-i\frac{\pi}{4}\sigma^L_y\right)$$

was performed. As in the DFS memory experiments, gradients were used to induce a spatially incoherent error over different parts of the sample. However, the noise effects associated with a time-independent gradient Hamiltonian (i.e. an infinitely long correlation time) would tend to be effectively averaged out by the applied control sequences as described by coherent averaging [31] and dynamical decoupling [23]. In order to make sure that the net action of the applied gradients is maintained in the presence of the external control, a procedure similar to the fast-switching control schemes discussed in [24] was followed, by rapidly modulating the strength of the applied gradient Hamiltonians over the course of the control sequence. Thus, a temporal incoherence was also superimposed at each spatial location in the sample, enforcing a finite correlation time $\tau_c$ (hence a non-zero cut-off frequency) in the spectral density describing the noise. In practice, the gradient waveform was determined via a random walk process, whose shape is depicted in figure 4. The gradient strength was changed every $50.6 \mu s$ therefore $\tau_c \sim 50.6 \mu s$. By making sure that $\tau_c$ is short compared with the control cycle time of the sequences used to implement the composite rotations ($\sim 700 \mu s$ in our case), active control is made ineffective at averaging out the high-frequency effects of the noise during the computation.

A broad range of values for the maximum applied gradient strength were explored to test the robustness of the computation to collective phase errors. The experimentally determined
Figure 4. The temporal variation of the gradient waveform used to test control of a DFS qubit in the presence of noise. The shape was determined by a random walk algorithm. The stepping time was $50.6 \mu s$, which is faster than any of the control timescales relevant in implementing the encoded transformations.

Figure 5. Experimentally determined entanglement fidelity for the implementation of a composite encoded $y$ rotation of $\pi/2$ in the presence of noise. Magnetic field gradients implement a spatially incoherent collective phase error as a function of the molecular position. Gradient strengths from 0 to $\sim 100$ kHz cm$^{-1}$ were applied over a 1 cm sample. The behaviour of $F_e$ is flat over a broad range of noise strengths and remains significantly above the 0.5 threshold for all noise values considered. This convincingly demonstrates the ability to control a DFS qubit in the presence of noise significantly stronger than the natural noise of the system.
gate entanglement fidelities are shown in figure 5. As expected, the computation is protected up to a particular noise intensity and then falls off with increasing noise strength. As in the memory case, it is worth stressing that \( F_e \) values well exceeding the value 0.50 have been achieved over the entire range of applied noise strengths. It should be noted that because the active sample is of the order of 1 cm, most of the sample is experiencing noise strengths significantly stronger than natural fluctuations—which are approximately 1 Hz in strength.

7. Conclusions

We have provided the first demonstration of universal control over a DFS-encoded qubit. The implementation relied on combining the benefits of passive noise protection via DFS coding with the ability of relaxing the constraints of fully DF manipulations via appropriate control design—thereby also validating the underlying principles of encoded decoupling. We believe that our techniques are applicable to a wide class of quantum information devices, where collective dephasing mechanisms play a dominant role and where the structure of the system’s internal Hamiltonian can be mapped onto an NMR-type Hamiltonian. These may include various solid-state proposals as discussed in [20, 34]. Thus, our results improve the prospects that DFS/NS coding, combined with encoded dynamical decoupling and robust control design, will play a practical role for both protected storage and manipulation of quantum information in QIP.

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References

[1] Nielsen M A and Chuang I L 2000 Quantum Computation and Quantum Information (Cambridge: Cambridge University Press)
[2] See also Zurek W H 2001 Decoherence, einselection, and the quantum origins of the classical Preprint quant-ph/0105127
[3] Zanardi P and Rasetti M 1997 Noiseless quantum codes Phys. Rev. Lett. 79 3306
Zanardi P and Rasetti M 1997 Error avoiding quantum codes Mod. Phys. Lett. B 11 1085
[4] Duan L-M and Guo G-C 1997 Preserving coherence in quantum computation by pairing quantum bits Phys. Rev. Lett. 79 1953
[5] Lidar D A, Chuang I L and Whaley K B 1998 Decoherence-free subspaces for quantum computation Phys. Rev. Lett. 81 2594
[6] Zanardi P 1998 Dissipation and decoherence in a quantum register Phys. Rev. A 57 3276
Zanardi P and Rossi F 1998 Quantum information in semiconductors: noiseless encoding in a quantum-dot array Phys. Rev. Lett. 81 4752
[7] Lidar D A, Bacon D and Whaley K B 1999 Concatenating decoherence-free subspaces with quantum error-correcting codes Phys. Rev. Lett. 82 4556
[8] Bacon D, Lidar D A and Whaley K B 1999 Robustness of decoherence-free subspaces for quantum computation Phys. Rev. A 60 1944

[9] Lidar D A, Bacon D, Kempe J and Whaley K B 2001 Decoherence-free subspaces for multiple-qubit errors. I. Characterization Phys. Rev. A 63 022306

[10] Knill E, Laflamme R and Viola L 2000 Theory of quantum error correction for general noise Phys. Rev. Lett. 84 2525

[11] Viola L, Knill E and Lloyd S 2000 Dynamical generation of noiseless quantum subsystems Phys. Rev. Lett. 85 3520

[12] Viola L, Knill E and Laflamme R 2001 Constructing qubits in physical systems J. Phys. A: Math. Gen. 34 7067

[13] De Filippo S 2000 Quantum computation using decoherence-free states of the physical operator algebra Phys. Rev. A 62 052307

[14] Zanardi P 2001 Stabilizing quantum information Phys. Rev. A 63 012301

[15] Kwiat P G, Berglund A J, Altepeter J B and White A G 2000 Experimental verification of decoherence-free subspaces Science 290 498

[16] Zanardi D et al 2001 A decoherence-free quantum memory using trapped ions Science 291 1013

[17] Viola L, Fortunato E M, Pravia M A, Knill E, Laflamme R and Cory D G 2001 Experimental realization of noiseless subsystems for quantum information processing Science 293 2059

[18] Zanardi P 1999 Computation on an error-avoiding quantum code and symmetrization Phys. Rev. A 60 R729

[19] Bacon D, Kempe J, Lidar D A and Whaley K B 2000 Universal fault-tolerant quantum computation on decoherence-free subspaces Phys. Rev. Lett. 85 1758

[20] Loss D and DiVincenzo D P 1998 Quantum computation with quantum dots Phys. Rev. A 57 120

[21] Kane B 1998 A silicon-based nuclear spin quantum computer Nature 393 133

[22] Platzman P M and Dykman M I 2000 Quantum computing with electrons floating on liquid helium Science 284 1967

[23] Havel T F, Sharf Y, Viola L and Cory D G 2001 Hadamard products of product operators and the design of gradient-diffusion experiments for simulating decoherence by NMR spectroscopy Phys. Lett. A 280 282

[24] Hall L D and Norwood T J 1986 Zero-quantum-coherence, chemical-shift-resolved imaging in an inhomogeneous magnetic field J. Magn. Res. 67 382

[25] Ernst R R, Bodenhausen G and Wokaun A 1994 Principles of Nuclear Magnetic Resonance in One and Two Dimensions (Oxford: Oxford University Press)

[26] Kraus K 1983 States, Effects, and Operations (New York: Springer)

[27] Warren W S et al 1998 MR imaging contrast enhancement based on intermolecular zero quantum coherences Science 281 247

[28] Warren W S et al 1996 Homogeneous NMR spectra in inhomogeneous fields Science 272 92

[29] Vathyam S, Lee S and Warren W S 1996 Homogeneous NMR spectra in inhomogeneous fields Science 272 92

[30] Lin Y Y, Ahn S, Murali N, Brey W, Bowers C R and Warren W S 2000 High-resolution, >1 GHz NMR in
unstable magnetic fields *Phys. Rev. Lett.* **85** 3732

[30] Fortunato E M, Pravia M A, Boulant N, Teklemariam G, Havel T F and Cory D G 2002 Design of strongly modulating pulses to implement precise effective Hamiltonians in quantum information processing *J. Chem. Phys.* to be published (Preprint quant-ph/0202065)

[31] Haeberlen U and Waugh J S 1968 Coherent averaging effects in magnetic resonance *Phys. Rev.* **175** 453

[32] Haeberlen U (ed) 1976 The corrections arising within AHT from finite pulse widths have been analyzed, for instance *High Resolution NMR in Solids: Selective Averaging* (New York: Academic)

[33] Viola L 2001 On quantum control via encoded dynamical decoupling Preprint quant-ph/0111167

[34] Lidar D A and Wu L-A 2001 Reducing constraints on quantum computer design by encoded selective recoupling 2001 *Phys. Rev. Lett.* **88** 017905

[35] Hahn E L 1950 Spin echoes *Phys. Rev.* **80** 580

[36] Shaka A J and Freeman R 1983 Composite pulses with dual compensation *J. Magn. Res.* **55** 487

[37] Sodickson A and Cory D G 1998 A generalized $k$-space formalism for treating the spatial aspects of a variety of NMR experiments *Prog. Nucl. Magn. Reson. Spectrosc.* **33** 77

[38] Chuang I L, Gershenfeld N, Kubinec M G, Leung D W 1998 Bulk quantum computation with nuclear magnetic resonance: theory and experiment *Proc. R. Soc. A* **454** 447

[39] Chuang I L and Nielsen M A 1997 Prescription for experimental determination of the dynamics of a quantum black box *J. Mod. Opt.* **44** 2455

[40] Schumacher B 1996 Sending entanglement through noisy quantum channels *Phys. Rev. A* **54** 2614

[41] Bowdrey M D, Oi D K L, Short A J and Jones J A 2001 Fidelity of single qubit maps Preprint quant-ph/0103090

[42] Bennett C H, DiVincenzo D P, Smolin J A and Wootters W K 1996 Mixed-state entanglement and quantum error correction *Phys. Rev. A* **54** 3824

[43] Freeman R, Wittekoek S and Ernst R R 1970 High-resolution NMR study of relaxation mechanisms in a two-spin system *J. Chem. Phys.* **52** 1529

[44] Hartman S R and Hahn E L 1962 Nuclear double resonance in the rotating frame *Phys. Rev.* **128** 2042

[45] Chingas G C, Garroway A N, Bertrand R D and Moniz W B 1981 Zero quantum NMR in the rotating frame: $J$-cross-polarization in $AXN$ systems *J. Chem. Phys.* **74** 127

[46] Meiboom S and Gill D 1958 Modified spin-echo method for measuring nuclear relaxation times *Rev. Sci. Instrum.* **29** 688

[47] Freeman R, Kempsell S P and Levitt M H 1980 Radio-frequency pulse sequences which compensate their own imperfections *J. Magn. Res.* **38** 453

[48] Shaka A J, Keeler J and Freeman R 1983 Evaluation of a new broad-band decoupling sequence: WALTZ 16 *J. Magn. Res.* **53** 313