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Nuclear spin qubits in a pseudospin quantum chain

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We analyze a quantum-computer design based on nuclear spin qubits in a quasi-one-dimensional chain of non-Kramers doublet atoms. We explore the use of spatial symmetry breaking to obtain control over the local dynamics of a qubit. We also study the decoherence mechanisms at the single qubit level and the interactions mediated by the magnetic media. The design can be realized in $\mathrm{PrBr}_{3-x}\mathrm{F}_x$ with nuclear magnetic resonance techniques.

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I. INTRODUCTION

Nuclear magnetic resonance (NMR) is the framework of a very promising quantum-computing architecture [1]. NMR is a natural choice because nuclei are protected from many sources of decoherence, and therefore produce robust qubits. Successful realizations of quantum algorithms implemented on a NMR quantum computer (QC) have been realized in liquid solutions of molecules [2,3]. Nevertheless, a liquid NMR QC is not easily scalable, that is, there is a practical limit in the number of qubits that can be constructed in a molecule. From a handful of qubits already achieved one must scale the QC to several thousands before a nontrivial algorithm can be run [2,3]. Though other limitations can also be argued to the use of NMR [4], scalability is an undeniable problem.

A possible route to deal with the scalability problem is to consider NMR in crystals [5]. There are several different proposed designs, but all of them share two common elements. First, a gradient magnetic field is used to shift the nuclear resonance frequencies of different nuclei, allowing qubits to be addressed independently. Secondly, as the number of qubits increases, a second decoherence channel is introduced by the low energy excitations of the interacting qubits. In any crystal, the direct dipolar interaction between nuclei produces secular broadening. To a certain extent this broadening can be reduced by NMR techniques. Thus, it is usually assumed that a perfect selective decoupling of the qubits from the dipolar interaction can be achieved.

Although very promising, there are technical problems with the use of NMR in crystals. For example, in the proposed materials CaF_2 and MnF_2, qubits are the nuclear spin $1/2$ of the F ions [5]. To obtain a measurable frequency shift from one qubit to another a homogeneous gradient field of more than $1\ \mathrm{T}/\mu\mathrm{m}$ is required. The obvious solution is to separate qubits from each other. However, by distancing the qubits to work with an experimentally feasible value of the field gradient, another problem is created by weakening the qubit-qubit interactions.

Interacting qubits are a necessary condition for quantum computation. A quantum algorithm is a sequence of unitary transformations in the Hilbert space spanned by all the qubits. A given transformation in a subspace of n qubits is called a n-qubit gate. A quantum-computing scheme must provide a complete set of such quantum gates, in other words, it must be possible to construct any unitary transformation with a sequence of building block operations provided by the design. One of the most useful results in quantum-information theory is that from all one-qubit gates and almost any two-qubit gate is possible to find a complete set of gates [6]. In a NMR QC, the one-qubit gates are easily produced. The two-qubit gate is the time evolution of two qubits under an interaction.

The viability of a solid state NMR QC relies on interactions available to construct the two-qubit gate and the correspondent decoherence times. On the one hand, in CaF_2, the only available interaction is the direct dipolar coupling between nuclear moments. In most cases this interaction is effectively short ranged for quantum-computational purposes. The small nuclear moments and the $1/r^3$ dependence makes the operation time of a gate (composed by two qubits far apart) much larger than the decoherence times. On the other hand, in MnF_2, the relevant interaction is the Suhl-Nakamura coupling [7]. This is an indirect coupling of nuclear spins mediated by magnons of the Mn electronic spins. Below its Néel temperature the magnon spectrum has a gap. At the same time that a gap reduces decoherence, it implies that the interaction strength has an exponential decay with the distance. Thus, it is unlikely that a considerable separation between qubits can be obtained in both cases. The search for long-range interactions has motivated several recent publications [8,9]. Unfortunately, long-range interactions are tied to low-energy modes and, consequently, short decoherence times.

In this paper, we discuss nuclear-nuclear interactions mediated by an anisotropic quantum-pseudo-spin chain. We analyze how the breaking of spatial symmetries in a system of non-Kramers ions can be used to gain control over local properties of a QC. We show that one can reduce decoherence and/or construct different two-qubit gates as a function of external electromagnetic fields. Although our ideas are general, we propose a specific realization in the compounds $\mathrm{PrCl}_{3-x}\mathrm{F}_x$ and $\mathrm{PrBr}_{3-x}\mathrm{F}_x$. Both materials are equally suitable to our discussion, but we use the parameters of the latter in our estimates. We start by summarizing the properties of the parent compound $x=0$. Subsequently, we discuss the chemical doping with F. Finally, we explore the use of the nuclear spin from the F ions as qubits.

II. THE PHYSICS OF PrBr_3 AND THE CONSTRUCTION OF QUBITS

PrBr_3 is a one-dimensional (1D) ionic insulator made out of Pr chains separated by 5 Å. The Pr ions are subjected to a

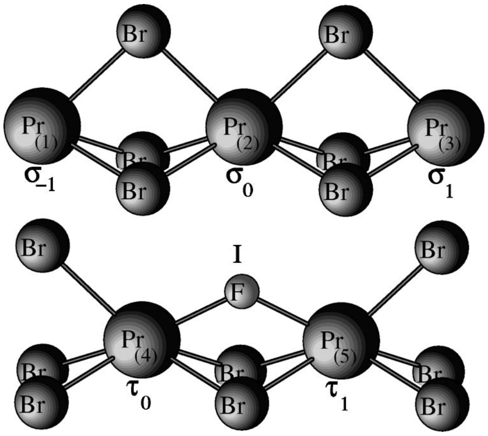

FIG. 1. Two adjacent chains in $\mathrm{PrBr}_{3-x}\mathrm{F}_x$.

crystal field with C_{3h} symmetry. Their ground state is a non-Kramers doublet that is separated from the first excited state by a gap of 17 K [10]. A Jahn-Teller transition takes place at 0.1 K [11], it lifts the doublet degeneracy, and sets a low-temperature limit to the applicability of this material to our design. A convenient way to model this system is via a pseudo-spin-1/2 representation [12]. We focus on the physics of two adjacent chains and we label the pseudospins of each of these chains as τ^z and σ^z (see Fig. 1). The single ion Hamiltonian at site i is written as

$$H_{\mathrm{ion}} = \sum_i \hbar \gamma_s^z B_z S_i^z + g_s^x E_x S_i^x + g_s^y E_y S_i^y ,$$

where $\vec{S}=\{\vec{\sigma},\vec{\tau}\}$, $\gamma_s^z = 1.4\times 10^{11}\ \mathrm{T}^{-1}\ \mathrm{s}^{-1}$, $\vec{B}$ is an external magnetic field and $\vec{E}$ an applied electric field [10]. We are unaware of published values for the electric dipolar constants in PrBr_3, however, they should not be very different from the ones in PrCl_3 where $g_s^{x,y}=4.0\times 10^{-31}$ C m [13]. It is important to stress that there is no off-diagonal matrix element that couples the doublet state to the magnetic field. Therefore, a magnetic field cannot induce transitions between the doublet states. The ionic magnetic moments are coupled by a dipolar term, however the most relevant contribution to the interaction Hamiltonian comes from transitions due to the transverse electric dipoles that are strongly coupled to the lattice. Although the only real magnetic moment is oriented along the chain (z direction), this family of compounds is regarded as XY chains described by the Hamiltonian

$$H_{xy} = J_\perp \sum_i S_i^x S_{i+1}^x + S_i^y S_{i+1}^y , \tag{1}$$

where $J_\perp \approxeq 3$ K [14].

In order to construct a qubit, we propose the use of the nuclear spins of F ions in the diluted salt $\mathrm{PrBr}_{3-x}\mathrm{F}_x$. There are two main components to nuclear decoherence, connected with the two strongest interactions that a nucleus is subjected to the electric quadrupolar and the magnetic dipolar [15]. We are ultimately interested in the decoherence channels in a F nucleus in $\mathrm{PrBr}_{3-x}\mathrm{F}_x$. Nuclear quadrupole resonance experiments have measured $T_{1,2}$ for the Br nuclei in the parent compound. They established fairly well that the spin-lattice relaxation time T_1 is due to magnetic interactions [14,16] and it is of order of 100 ms at 1 K. Moreover, the nuclear spin-spin decoherence time T_2 was found to be $\approxeq 40\ \mu$s at 1 K. The decoherence sources that lead to this value for T_2 are not yet well understood [11]. If we use the Van Vleck formula [15] to estimate the secular broadening of resonance lines, we find that the direct dipolar interaction among the nuclei leads to a broadening of the order of $10^2\ \mu$s. Further considering the quadrupolar effects it is clear that the direct dipolar interaction gives a sizable contribution to decoherence. Thus, as usual in solid state NMR designs, we can conclude that decoupling is very important in order to make this family of compounds useful to a QC.

Each F introduces a local lattice distortion, hence lowering the crystal field symmetry at neighboring Pr ions. The distortion introduced by the F ion has its strongest effect on the Pr ions labeled 2, 4, 5 in Fig. 1. In the pseudospin representation, a local symmetry breaking corresponds to the addition of transverse fields $\overline{\Delta}$ and Δ on each one of these sites. Moreover, the $\mathrm{Pr}_{(4,5)}$ no longer have a plane of inversion perpendicular to the chain axis. Thus, these ions can develop electric dipoles perpendicular to that plane. The Hamiltonian for the pseudospin chains can be written as

$$H_{\mathrm{Pr}} = H_{\mathrm{ion}} + H_{xy} + \Delta\sigma_0^x + \overline{\Delta}(\tau_0^x + \tau_1^x) + g_s^z E_z(\tau_0^z - \tau_1^z) . \tag{2}$$

We consider the case where $(\overline{\Delta},\Delta) \ll \max(\hbar\gamma_s^z B_z, k_B T) < J_\perp$, otherwise the moments at $\mathrm{Pr}_{(2,4,5)}$ would be completely quenched by the symmetry breaking and the analysis below would need to be extended to include next-near-neighbor interactions. Notice that in Eq. (2) the transverse fields introduce matrix elements between the two magnetic states of $\mathrm{Pr}_{(2,4,5)}$. Thus, an oscillating magnetic field parallel to the chain axis would reveal two distinct resonant lines $\omega_{\overline{\Delta}}$ and ω_Δ associated with the splitting of the Pr doublet state.

III. THE QUBIT HAMILTONIAN

The use of F as a qubit has two advantages. There is no decoherence due to electric field gradients because it does not have a quadrupolar moment. In addition, there is only one isotope of F in nature, so all qubits experiencing the same magnetic field are identical. By assuming perfect decoupling, we can disregard the direct dipolar interaction between nuclei. This is a much less stringent condition than in other NMR QC schemes because the qubit resonance frequency is very distinct from the other ions. Therefore, straightforward pulse sequences can be used to perform the decoupling. The remaining contribution to the nuclear Hamiltonian comes from the magnetism of the surrounding Pr atoms. Hence, the nuclear hyperfine interaction of each F ion in first approximation can be written as

$$\begin{aligned} H_{\mathrm{F}} = {} & \left[\hbar\gamma_N B_z + d\left(\sigma_0^z - \frac{\tau_0^z + \tau_1^z}{2}\right) - \tilde{d}(\sigma_{-1}^z + \sigma_1^z)\right] I^z \\ & + 3d(\tau_0^z - \tau_1^z) I^x + \sqrt{2}\tilde{d}(\sigma_{-1}^z - \sigma_1^z) I^y , \end{aligned} \tag{3}$$

where $d=(\mu_0^2\hbar\gamma_s^z\gamma_N)/(4\pi r_0^3)\approxeq 10^{-4}$ K, $\tilde{d}\approx d/5$, $\gamma_N=25$

$\times 10^7\ \mathrm{T}^{-1}\ \mathrm{s}^{-1}$, and $\vec{I}$ is the nuclear spin-1/2 operator of the F nucleus.

The pseudospin physics described by Eq. (2) presents us with a very interesting situation. An applied magnetic field with frequency $\omega_{\overline{\Delta}}$ and/or an electric field E^z only affect the $\mathrm{Pr}_{(4,5)}$, and therefore can be used to act locally in the qubit. For example, a sufficiently large electric field forces τ_0 and τ_1 into a singlet configuration, freezing their dynamics. The net result is decoupling of the F ion from the τ chain. In this case the hyperfine Hamiltonian simplifies to

$$H_{\mathrm{F}} \approxeq [\hbar\gamma_N B_z + d\sigma_0^z - \tilde{d}(\sigma_{-1}^z + \sigma_1^z)]I^z + \sqrt{2}\tilde{d}(\sigma_{-1}^z - \sigma_1^z)I^y. \tag{4}$$

This is a particularly interesting effect. It cancels the strongest transverse part of Eq. (3), and consequently, corresponds to a reduction in the dissipation rates $T_{1,2}^{-1}$.

A. Dissipation rates

In order to estimate the dissipation rates due to the Pr magnetic moments, we will focus on the low-energy physics of Eq. (2). Therefore, we can use Abelian bosonization [17] to obtain simple analytical expressions for $T_{1,2}^{-1}$.

Bosonization is a well stablished method to study spin chains. In a concise way, we first use the Jordan-Wigner transformation, mapping the pseudospins in spinless fermions. Then, we linearizing the dispersion relations around the two Fermi points, $p_F = \arccos(\hbar\gamma_s^z B_z/J_\perp)$, and define the Fermi velocity $v = J_\perp \sin(p_F)$. The result is that H_{xy} can be rewritten as a free bosonic Hamiltonian. In this language, it is straighforward to evaluate the pseudospin correlation function at zero temperature [17]

$$\langle S_j^z(\tau) S_0^z(0)\rangle = \frac{1}{2\pi^2}\frac{x^2 - (v\tau)^2}{[x^2 + (v\tau)^2]^2}\frac{\cos(2p_K x)}{2\pi^2}\frac{1}{x^2 + (v\tau)^2}, \tag{5}$$

where τ is the imaginary time, $x = a_0 j$, and $a_0 \approx 4.4$ Å is the lattice spacing.

For a sufficiently large magnetic field ($B_z \gg 0.1$ T), T_1^{-1} is given by [18]

$$T_1^{-1} = \frac{1}{2}\int_{-\infty}^{\infty} dt' \langle H_\perp(t) H_\perp(t+t')\rangle e^{-i\omega_0 t'}, \tag{6}$$

where, if we focus in the regime described by Eq. (4), we defined

$$\omega_0 = \gamma_N B_z,$$

$$H_\perp = \sqrt{2}\tilde{d}(\sigma_{-1}^z - \sigma_1^z).$$

An equivalent expression for T_2 is obtained when we match the results of a random phase approximation (RPA) calculation for the transverse suceptibility with the solution of the Boch's equations [19]. Using Eq. (5) in Eq. (6) or the RPA result, we evaluate the zero-temperature decoherence rates due to the pseudospins as

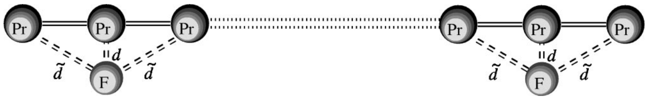

FIG. 2. Interaction between two nuclear spins of F ions is mediated by the pseudospin chain of Pr. d and $\tilde{d}$ are the strength of the hyperfine coupling defined in Eq. (3).

$$T_1^{-1} = T_2^{-1} \approxeq 8\pi^{-1}\gamma_N(\hbar\tilde{d}\gamma_s^z)^2 B_z^3 J_\perp^{-4}. \tag{7}$$

The unusual dependence of the relaxation time with the magnetic field, scaling as B_z^3, can be used to assert Eqs. (3) and (4). Finally, an applied transverse electric field ($E_{x,y} \neq 0$) can be used to open a gap in the pseudospin spectrum. This further isolates the qubit by quenching the pseudospins magnetic moments, and therefore, even smaller values of $T_{1,2}^{-1}$ can be achieved.

In general, nuclear spins interacting with a gapless spin chain would have super-ohmic dissipation. However, the hyperfine Hamiltonian, (4) that we derive depends exclusively on the z component of the pseudospins. This restricted dipolar interaction implies an ohmic dissipation. We emphasize that this is somewhat unique feature of pseudospins. If Eq. (4) would have flip-flop terms, then the transverse correlations of the spseudo spins would imply a super-ohmic behavior.

B. Construction of quantum gates

Now that we have studied the single qubit problem, we turn our attention to the qubit-qubit interaction. We focus in the regime described by Eq. (4) because it is the most favorable for QC. Consider a second F atom along the chain as shown in Fig. 2. By integrating out the σ spins we obtain a retarded interaction between the two nuclei. This is very similar to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, but mediated by the pseudospins [20].

Exactally as in the RKKY problem, the F nuclear spins have a much slower dynamics than the pseudospins ($\gamma_N B_z \ll J_\perp/\hbar$). Therefore, it is reasonable to consider an instantaneous approximation to the interaction. At zero temperature, we use Eq. (5) to calculate its form.

For the RKKY, finite-temperature corrections are usually irrelevant because the Fermi energy is much larger than the temperatures under consideration. However, in the pseudospin chain we are assuming temperatures only one order of magnitude smaller than $J_\perp$. We can easily rewrite the zero-temperature correlation function (5) in its finite-temperature form by using the conformal invariance of the XY model [21]. The final result is the effective interaction between to qubits

$$H_{\mathrm{eff}} \approxeq f_{zz} I_1^z I_2^z + f_{yz}(I_1^y I_2^z + I_1^z I_2^y) + f_{yy} I_1^y I_2^y, \tag{8}$$

where we have defined the nuclear exchange couplings

$$f_{zz} = d^2 G(\Delta x) - d\tilde{d}[G(\Delta x - 1) + G(\Delta x + 1)]$$
$$+ \tilde{d}^2[2G(\Delta x) + G(\Delta x + 2) + G(\Delta x - 2)],$$

$$f_{yz} = \sqrt{2}\{d\tilde{d}[G(\Delta x - 1) - G(\Delta x + 1)] + \tilde{d}^{\,2}[G(\Delta x + 2) - G(\Delta x - 2)]\},$$

$$f_{yy} = 2\tilde{d}^{\,2}[2G(\Delta x) - G(\Delta x + 2) - G(\Delta x - 2)]. \qquad (9)$$

$G(\Delta x)$ is the finite-temperature pseudospin propagator given by

$$G(\Delta x) \approxeq \frac{1 - \cos(2p_F\Delta x)}{2\pi^2 v^3\beta^2}\left[\sinh\left(\frac{\Delta x}{v\beta}\right)\right]^{-2}, \qquad (10)$$

where Δx is the distance between qubits in units of lattice spacing a_0 and $\beta = 1/(k_B T)$. For distances smaller than the thermal coherence length $\xi_T = v\beta$, the interaction decays as a power law $G(\Delta x) \approxeq (1 - \cos[2p_F\Delta x])/(2\pi^2 v\Delta x^2)$, leading to long-range interaction between qubits. It is also interesting to consider the consequences of applying transverse electric fields. Since the pseudospin propagator acquires a gap, there is an additional exponential decay in Eq. (8) which is a function of $E_{(x,y)}$. Thus, we can use transverse fields to switch on and off the interaction between qubits.

Equation (8) is a two-qubit gate. In conjunction with the possibility to perform arbitrary rotations, it generates a complete set of quantum gates [6]. The inverse of the gate operation time is given by $T_G^{-1}(\Delta x) = \hbar^{-1}\min(|f_{zz}|, |f_{yz}|)$. In order to compare T_G with $T_{1,2}$ we consider a particular case. Take $B_z \approx 2$ T and a temperature $T = 0.1$ K, so that the pseudospin chain is partially polarized. Low-temperature corrections to Eq. (7) are very small, and we use it as an upper bound estimate to the decoherence times $T_{1,2}^{-1} \sim 10^{-2}$ s^{-1}. These values are much smaller than the rates in PrBr$_3$ due to three facts: the absence of quadrupolar effects, the reduction of pseudospin fluctuation in $\tau_{0,1}$ and the assumption of decoupling. Two qubits separated by 13 Å have $T_G(3) \sim 10^{-1}$ s, thus leading to a quantum gate at the edge of the error correction threshold of 10^{-4} [3,22].

Another important aspect of Eqs. (2) and (3) is that several different gates can be constructed as a function of the magnetic field B_z, the resonance frequencies $\omega_{\Delta,\bar{\Delta}}$ and the electric fields $E_{x,z}$. For instance, the pseudospin propagator (10) has an oscillatory behavior with B_z. This can be used to change the relative strength of f_{ij} in Eq. (8). In order to make this point clear, we now pause and consider a concrete example.

One of the most simple quantum circuits is the one that creates entangle pairs of qubits (Bell's states). From the quantum-logic perspective, this is accomplished by the use of a Hadamard gate followed by a controlled-NOT (CNOT) gate [23]. Since the production of entangle pairs is fundamental to perform quantum computation and quantum comunication, this straightforward circuit is a conerstone in any design. The key element here is the CNOT gate. It is a two qubit gate and, consequently its implementation depends upon the avaiable interaction. In liquid state NMR the strongest component in the Hamiltonian that a pair of qubits is subjected is [3]

$$H_z \cong J I_1^z I_2^z. \qquad (11)$$

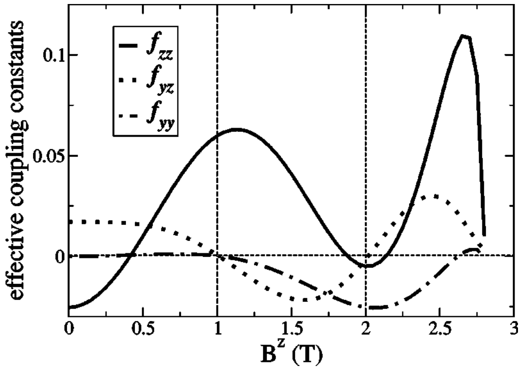

FIG. 3. The coupling constants f_{zz}, f_{yz}, and f_{yy} as a function of the magnetic field B^z in units of $\tilde{d}^2$ for two F atoms separated by four lattice spacing.

This Hamiltonian can also be approximated by Eq. (8). For the sake of argument, let us assume two F atoms separated by four lattice sites ($\sim$18 Å). In addition, let us consider the external conditions that we considered before: a large E^z to freeze the pseudospin dynamics in $\tau_{0,1}$ and $T = 0.1$ K.

From Eq. (8) and the definition of the pseudospin propagator we can plot Fig. 3, where we can see that for $B^z \cong 1.01$ T the effective coupling constants are

$$\frac{f_{zz}}{\tilde{d}^{\,2}} \cong 0.06,$$

$$\frac{f_{yz}}{\tilde{d}^{\,2}} \cong 0.001,$$

$$\frac{f_{yy}}{\tilde{d}^{\,2}} \cong 0.001.$$

Thus, as in liquid NMR, the strongest component in the interaction is given by Eq. (11). In order to produce a CNOT gate with this Hamiltonian in an NMR setup [3], one must first apply a radio frequency pulse to rotate I_2 about $\hat{x}$ ($+\hat{z}$ goes to $-\hat{y}$). Then the spin system evolves with Eq. (11) for a time $t = \pi\hbar/4J$. Then, a second pulse is sent to rotate I_2 by 90° about the $-\hat{y}$ axis. Finally, an additional phase shift on both spins is used to obtain the CNOT gate. If we add the initial Hadamard gate, it is necessary to use a total of five radio frequency pulses (one-qubit gates) and the time evolution of the Hamiltonian (11).

Let us analyze another possibility. Consider the same condictions as before, but with an external magnetic field $B^z = 2.02$ T. In this case, the effective coupling constants are

$$\frac{f_{zz}}{\tilde{d}^{\,2}} \cong -0.005,$$

$$\frac{f_{yz}}{\tilde{d}^{\,2}} \cong 0.0003,$$

$$\frac{f_{yy}}{\tilde{d}^2} \cong -0.025.$$

In contrast with the previous case, the strongest part of the interaction is

$$H_y \cong f_{yy} I_1^y I_2^y. \tag{12}$$

If we allow a free evolution of the system by Eq. (12) for a time $t = \pi\hbar/2f_{yy}$, the unitary transformation that is implemented is

$$R = \begin{bmatrix} 1 & 0 & 0 & -i \\ 0 & 1 & i & 0 \\ 0 & i & 1 & 0 \\ -i & 0 & 0 & 1 \end{bmatrix}.$$

Acting on the computational basis with this rotation we automatically generate the entangled states

$$|\beta_1\rangle = \frac{\sqrt{2}}{2}(|00\rangle - i|11\rangle),$$

$$|\beta_2\rangle = \frac{\sqrt{2}}{2}(|01\rangle + i|10\rangle),$$

$$|\beta_3\rangle = \frac{\sqrt{2}}{2}(|01\rangle - i|10\rangle),$$

$$|\beta_4\rangle = \frac{\sqrt{2}}{2}(|00\rangle + i|11\rangle).$$

Hence, one can fine tune the experimental setup to obtain a desired quantum circuit using less resources. In the above example, the simple tuning of the magnetic field replace the one qubit gates on the previous setting. However, this is just one of many possible ways to control the interaction Hamiltonian. A more subtle (and potentially more interesting way) is related to the frequency ω_Δ and $\omega_{\bar{\Delta}}$. In presence of a gradient magnetic field they have a site index ($\hbar\omega_\Delta \cong \sqrt{[\hbar\gamma_s^z B^z(\vec{x})]^2 + \Delta^2}$). Thus, one could act in the magnetic environment of each individual qubit.

IV. DISCUSSION AND CONCLUSIONS

Until this point we discussed how single qubits can be constructed and how a pair of qubits can interact. We now discuss how to use these building blocks in a QC.

The natural geometry is to consider a magnetic field gradient applied along the chain direction. Nuclei in the same equipotential line belong to different copies of the QC, and we assume that they can be periodically arranged (see below).

Initialization is a very hard problem in QC's based on nuclear spin qubits. However, there are some possible solutions already available in the literature [9,24]. At first sight one could imagine that the initialization could be done by optical pumping (Pound-Overhauser effect) with the pseudospins, as it is done in MnF_2 with electronic spin. Unfortunately, the same property that gives a lower decoherence rate than in other gapless magnetic systems hinders this option. Since there is no flip-flop term (S^+I^-) in the hyperfine Hamiltonian, one cannot use the pseudospins to pump the nuclear spins. There are two other possible "hardware" solutions that can be used to solve the initialization problem. A diluted set of magnetic impurities can be used to refrigerate the qubits. The general idea is to add a small amount of an ion with a large magnetic moment (such as Gd replacing some Pr) to the sample. This set of impurities can be used to pump energy out of the nuclear systems and after some polarization is achieved a sufficiently large magnetic field would "freeze" the impurities. There are two setbacks in this approach. First, the Gd ion would "break" the pseudospin chains and the F ions in each side might not interact. Secondly, virtual flips of the Gd spin could introduce an additional decoherence channel. The second "hardware" solution is based on the fact that the crystals can be grown on a semiconductor substrate. By exciting the electron gas in the semiconductor, it is possible to use "cross-polarization-coherent transfer techniques." The latter is the solution found in Ref. [9] to the initialization procedure in a QC based on 1D organic molecules. Finally, if only partial polarization is obtained by one of the "hardware" methods cited above, the Schulman-Vazirani procedure [24] can be used as a "software" method to initialize the state.

The final element in a QC design is the read-out mechanism. All QC's based on NMR of impurities have the common problem of low signal due to the small density of qubit copies. However, nuclear polarization can increase considerably the NMR sensitivity. In this case, the read-out of a qubit with only 10^{12} copies is possible with current NMR technology [9].

There are some relevant experimental questions that are open and can foster new theoretical work. In the first place, the simplest way to produce crystals of a salt such as $PrBr_{3-x}F_x$ is through dehydration of a liquid solution [25]. This straightforward process creates samples with the F ions in random positions. Although this is sufficient to infer our results for a single qubit, further developments in ionic crystal growth should be accomplished before the full range of possibilities that we discuss can be experimentally studied. One possible research avenue is a molecular-beam epitaxial growth (MBEG). MBEG is a well established technique in semiconductors and metals. Although from a historical perspective the growth of ionic crystals is an old field, the technology is much less mature. Nevertheless, it shows unique characteristics that are worth exploring [26–28]. The most interesting feature is that the incoming molecule has a very weak bound with the surface terrace and strong bounding to the ledge. This can be simply understood in electrostatic terms, and as a consequence, leads to a large surface diffusion until the molecule reaches the ledge. We speculate that this fact can be used to obtain a higher degree of control in the impurity placement than in any other kind of material. Another interesting characteristic is that large lattice misfits are also allowed in the growth of layers. Thus, it is natural to propose experiments with a crystal composed of a superlattice of $PrBr_3$ and layers of $PrBr_2F$. This setup is feasible with the current technology and many of our results for the qubit-

qubit interaction can be experimentally tested. Another possibility is a super-lattice of $PrBr_3$ with layers of PrF_3, however, the large lattice misfit will probably prevent the layer growth [28]. A final remark is that ionic crystals grow well on semiconductors surfaces. This has two main consequences: (1) the semiconducting substrate can be integrated in other quantum computer schemes (similar to Si/P proposals) and with current electronics and (2) a semiconductor substrate can be used to initialize the quantum computer by optical pumping as we argued above.

There is another issue that is common to all solid state NMR designs: it is unlikely that perfect decoupling can be achieved. Therefore, the experimental value of T_2 is potentially smaller than the prediction of Eq. (7). Although we are probably overestimating T_2, we are also underestimating the gate time T_G. In order to derive Eq. (3), we assumed an specific form to the hyperfine interaction. Following the experimental results in $PrBr_3$ and $PrCl_3$, we assumed that the dipolar part is the most relevant component in the hyperfine Hamiltonian. This conclusion arises from the hypothesis that the chemical bound is truly ionic. In general there are some covalent components to the bound and this leads to a much stronger interaction with the electronic moments of adjacent ions. For instance, this is precisely what happens in MnF_2 [12,29]. Whereas our hypothesis is based on the experimental facts in $PrBr_3$ [11,14,16], a thorough experimental study should be done to assert the hyperfine Hamiltonian.

In summary, we showed how a non-Kramers ionic crystal has unique properties that can be exploited in a solid state NMR QC. We propose that chemical substitutions in such system can be used to encode quantum information and, at the same time, break the spatial symmetries. This controllable symmetry breaking can be used to act locally in the magnetic environment of the qubit, thus, having important consequences to decoherence and the construction of quantum gates. We based our discussion in a well known family of materials. However, the general principle that we put forward can be applied in a much broader context. In $PrBr_{3-x}F_x$, we showed that a QC based on our ideas is scalable, the decoherence rates are low, the interactions between qubits can be long ranged, and the qubits can be individually accessed with moderate magnetic field gradients.

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