Scalable designs for quantum computing with rare-earth-ion-doped crystals

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Due to inhomogeneous broadening, the absorption lines of rare-earth-ion dopants in crystals are many order of magnitudes wider than the homogeneous linewidths. Several ways have been proposed to use ions with different inhomogeneous shifts as qubit registers, and to perform gate operations between such registers by means of the static dipole coupling between the ions. In this paper we show that in order to implement high-fidelity quantum gate operations by means of the static dipole interaction, we require the participating ions to be strongly coupled, and that the density of such strongly coupled registers in general scales poorly with register size. Although this is critical to previous proposals which rely on a high density of functional registers, we describe architectures and preparation strategies that will allow scalable quantum computers based on rare-earth-ion-doped crystals.

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INTRODUCTION

Several proposals have been made for quantum computers based on rare-earth ions embedded in cryogenic crystals [1–3]. In the REQC (rare-earth quantum computing) proposals, qubits are identified with frequency channels appropriately selected within the inhomogeneous absorption profile of the dopant ions in the crystal. Full N-bit quantum computer instances occur at random in the material where a collection of ions with the selected absorption frequencies happen to be physically close enough to be coupled by the interaction between their excited state dipoles.

Due to their good coherence properties, rare-earth-ion-doped crystals constitute a promising solid state hardware testbed for quantum information experiments. Coherence times as long as 6.4 ms has been reported for the optical transition [4]. It has also been shown that the coherence times for the hyperfine levels can be extended by applying an appropriate magnetic field, and coherence times of 1.4 s have recently been demonstrated [5]. It has also been suggested that in some material the hyperfine coherence time could be extended to times on the order of several hundred seconds [5]. For these systems, coherent state preparation and quantum state tomography have been demonstrated for states connected by optical transitions [6]. Experiments have been performed to isolate and manipulate individual hyperfine transitions, which opens up the possibility to utilize the long coherence times of the hyperfine levels [7,8].

In this paper we discuss means to achieve scalable quantum computing in the rare-earth-doped crystals. In Sec. I, we briefly review the REQC idea for one-and two-bit gates in the rare-earth-ion quantum computer. In Sec. II, we discuss quantitatively the scaling problem, and we suggest to use a bus-architecture where a single ion is dedicated to provide the communication within the quantum computer instances and to ensure that there is with large probability precisely one ion present at each frequency channel. In Sec. III, we address the possibility of working with only a single instance of the quantum computer, rather than a large ensemble of identical instances. For this purpose it is necessary to have a single species with very high readout efficiency (single-ion detection). Section IV concludes the paper.

I. ONE- AND TWO-QUBIT GATES

In this section we briefly introduce the rare-earth quantum computing proposal, described in more detail in Refs. [1,8–11].

For the purpose of this paper, we consider rare-earth ions with three metastable ground state hyperfine levels, labeled $|0\rangle$, $|1\rangle$, and $|\mathrm{aux}\rangle$, which are all coupled by optical transitions to an excited state $|e\rangle$ [Fig. 1(a)]. The excited state $|e\rangle$ is inhomogeneously shifted by up to several GHz. Since the homogeneous linewidth of the optical transitions can be as low as kHz, this allows us to address a very large number of *channels*: frequency selected subgroups of the ions identified by their inhomogeneous shift [Fig. 1(b)]. Different channels are selected at sufficiently different frequencies to allow unambiguous addressing, and each channel is separated from the broad inhomogeneous profile by spectrally hole burning an interval around the frequency channel, so that only relatively narrow structures in frequency space are addressed by laser fields (Fig. 2). It is now possible to use the optical frequency to selectively perform quantum gates on the individual qubits, i.e., drive transitions between the qubit-levels $|0\rangle$ and $|1\rangle$ via the excited state $|e\rangle$.

Multiqubit gates are mediated by the interaction of the static dipole moment of the ions, which differs between the excited state $|e\rangle$ and the ground state hyperfine levels. The

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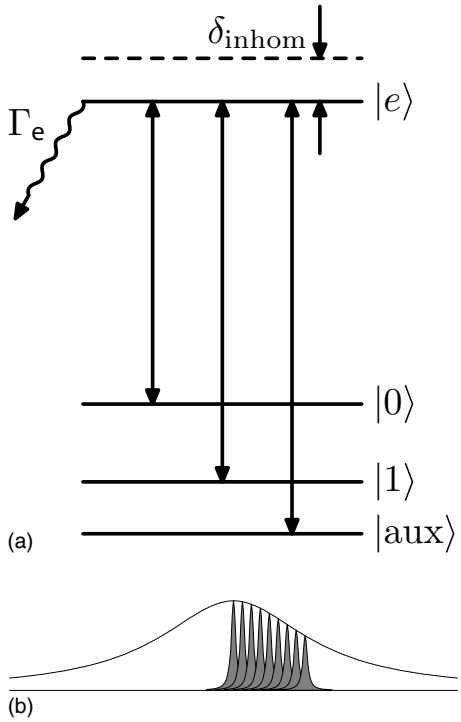

FIG. 1. Simplified level scheme for a crystal-embedded rare-earth ion (a). Vertical arrows indicate optical transitions between the ground state hyperfine levels ($|0\rangle$, $|1\rangle$, and $|\text{aux}\rangle$ and an excited state $|e\rangle$. The inhomogeneous shift, δ_{inhom}, of the $|e\rangle$-level leads to inhomogeneous broadening of the optical transitions when observed in an ensemble setting (b).

strength of the coupling is different for different ion pairs as it depends on the relative position of the ions. By using the dipole blockade effect [12], however, it is possible to implement reliable quantum gate operations that act independently of the precise value of the strength of the coupling between the involved ions, provided that this strength exceeds a certain threshold on the order of the intervals vacated by hole burning around the channels [1,9]. We will consequently refer to pairs of ions as being coupled or not depending on whether their coupling strength exceeds the threshold value.

The ions are only addressed according to their optical excitation frequencies, and there will in general be a very large number of ions having the same frequencies. This is a desired feature of the proposal, since the readout, as in the case of the nuclear magnetic resonance (NMR) quantum

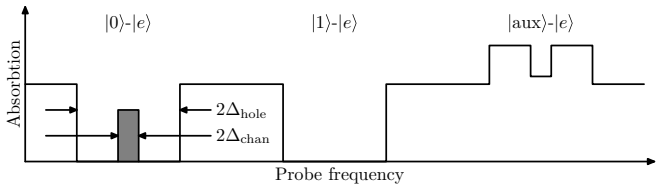

FIG. 2. Schematic structure of the channel absorption profile after the hole-burning procedure described in Refs. [1,9] has been applied. The ions absorbing within Δ_{hole} of the $|0\rangle - |e\rangle$ and $|1\rangle - |e\rangle$ transitions are ensured to have δ_{inhom} within Δ_{chan} of the channel center value and to be coupled above threshold to the desired controlling ions.

computer, is making use of the macroscopic signal from the entire ensemble. Unlike the NMR computer, however, the initial state is fully controlled, and all the ensemble members ideally perform the same unitary evolution. The readout is thus obtained from a pure state ensemble. In Refs. [10,11], it was further proposed to use composite pulses and suitably tailored continuous pulses to make the gates robust against the small frequency dispersion of the optical frequencies within the selected channels.

In weakly doped crystals like those used in recent experiments on Pr, Eu, and Tm doped crystals [6–8,13–15] the difference in dipole moment is sufficiently large that each qubit ion has several neighboring ions that it can control. Each of these neighbors can in turn control several other ions, etc. Consequently, from every ion there are branched chains of ions, where each ion in the chain can control its nearest neighbors. Thus, there is a large number of potential quantum processors in any crystal. Such a processor, however, operates with specific qubit frequencies located at random positions within the inhomogeneous absorption profile, and only if the experiment has the ability to detect single rare-earth ions, it will be possible to operate. Hence a good deal of theoretical and experimental work has focused on multiple instance implementations [6–11,13–17]. The multiple instance implementations are characterized by a choice of definite values for the qubit frequency channels followed by an optical pumping/hole burning to identify the instances where these selected frequency channels all are occupied by (at least) one ion.

II. ENSEMBLE QUANTUM COMPUTING WITH BUS IONS

A. Use of a bus ion for communication within a quantum computer instance

For a two-bit gate to be possible the ions must be interacting, i.e., they have to be within a critical distance R of each other. In the original proposal for computing with doped ions, any pair of ions were required to be within this distance, i.e., they had to be within a ball of diameter R. Without changing the physical implementation of the quantum computer, it is possible to increase the number of functioning instances, by assigning the role to one of the ions to take care of all communication in the instance so that two-bit gate operations between any pair of ions takes place using only their mutual interactions with the dedicated bus-ion [11]. Using the intermediate bus ion, all ions are only required to be within a distance of R from the bus, i.e., the ions have to be within a sphere of diameter $2R$, centered around the bus. This enlargement of the interaction volume by a factor 8 thus enlarges by a factor of approximately 8^{N-2} the number of N-bit quantum computer instances in the crystal ($N=2$ ions have to be within distance R of each other, and only for three ions and more can the ions have larger mutual distances).

B. Use of a bus ion for hole burning and initialization of a quantum computer instance

An instance of the quantum computer is only operational if all qubit channels are populated by a single ion: In the

original proposal, hole burning is applied both to define the frequency channels and to remove ions which do not have sufficient interactions with the other qubits, either because they are not physically close enough or simply because there is no ion at that frequency in the vicinity at all. In the bus-architecture just outlined, we must thus secure that every ion interacts with the bus, otherwise the whole instance will be nonoperational, and all ions should be removed by the hole-burning procedure: We know that the excited bus shifts the channel frequencies, and thus protects ions from hole burning, and therefore hole burning at each channel frequency in the presence of an excited bus will leave all good instances intact and only vacate the useless qubit channels. It is also possible to remove the bus itself if it is not coupled to representatives of all channels. A specific procedure to obtain this configuration is proposed in Ref. [1] and partly experimentally demonstrated in [8]. This procedure relies on repeatedly pumping unusable ions out of their qubit states, thus transferring them with certainty to their $|\text{aux}\rangle$-state. After these hole-burning operations we are left with a stable crystal which contains a number of disjunct, independent, and perfect quantum computer instances.

C. Scaling of the proposal

The ensemble REQC proposal relies on a large number of equivalent instances of the quantum processors in a single crystal, so that the individual qubits can be readout by the macroscopic absorption or dispersion properties of the crystal around the qubit frequencies. The number of qubits which can be represented in an ensemble REQC system is limited by the number of instances available of a given size. In this section, we discuss the scaling of the number of valid instances with the required register size N.

Denoting the number of representatives of channel i coupled to a given bus ion by (the stochastic variable) n_i, we distinguish between two regimes depending on the probability $P(n_i>0)$ that at least one representative of channel i is coupled to the bus ion: The weakly doped regime where $P(n_i>0)\ll 1$, as studied in the original proposal [1], and the strongly doped regime where $P(n_i>0)\approx 1$.

Modeling the scaling properties of the proposal is complicated by the fact that the inhomogeneous shift of the ions is also caused by the dipole interaction, resulting in a strong correlation between large inhomogeneous shift and nearness to other dopant ions [18]. Nevertheless, to illustrate the different scaling properties of the two doping regimes, we will for simplicity assume the n_i's to be independent and identically distributed. In this case, the probability P_N of a bus ion being coupled to N channels is seen to be related to $P_1 \equiv P(n_i>0)$ by

$$P_N = P_1^N \tag{1a}$$

$$= 1 - N(1-P_1) + O((1-P_1)^2). \tag{1b}$$

This implies that in the weakly doped regime, the number of available instances, which is proportional to P_N, decreases exponentially with increasing N, implying that this regime is not suitable for scalable quantum computing (1a). On the other hand, we see that in the strongly doped regime, the number of available instances only decreases linearly with N, allowing operation with a large number of qubits (1b).

There are two problems associated with operating in the strongly doped regime: One problem is finding materials that allow operation in this regime. A more technical problem is that in order to achieve $P(n_i>0)$ close to unity, the probability of having more than one channel representative coupled to a bus ion would in most scenarios be significant. This would either require modified gate implementations or an extra initialization step to inactivate the extraneous dopant ions.

1. Materials consideration for the highly doped regime

How many representatives of a channel are coupled to a given bus is influenced by two parameters? The ion concentration and the coupling strength required to perform gate operations.

In our above analysis, we have focused on the dipole-blockade type gates, relying on a very large coupling strength (some MHz) between ions, requiring the ions to be close and hence limiting the number of useful instances. As pointed out in [19], a gate capable of operating at lower interaction strengths would make ions from a much larger volume available for quantum gates, greatly enhancing the system scaling properties. Unfortunately, a gate operation compatible with lower coupling strengths will necessarily be slower, and in particular, have a higher integrated population of the decay-prone excited state. In the Appendix we prove that in order to maximally entangle a pair of ions with the excited state dipole-dipole interaction, the total population, $P_e^{(\text{tot})}$, in the excited state during the gate operation must obey $\int P_e^{(\text{tot})} dt > 2/g$, where g is the dipole coupling strength. This implies that in order to obtain a high fidelity gate operation, the interaction strength must be orders of magnitude larger than the excited state decay rate.

This result in turn implies that, in order to obtain $P(n_i>0)$ close to unity, a high dopant concentration is needed. In fact, we find that for the required coupling strengths, randomly doped material are not available with sufficient dopant density. An alternative approach would be to use stoichiometrically doped materials, which should make it possible to reach the highly doped regime [18,20].

2. Selecting unique channel representatives

To use a material with a high average number of dopant ions coupled to each bus ion, we need to deactivate all but one of the dopant ions coupled to each bus ion. In the following, we outline a method for achieving this for one channel: this procedure should be repeated sequentially for all channels.

The first step consists of preparing the bus ion in the superposition state $(|0\rangle+|1\rangle)/\sqrt{2}$. Secondly a phase shift of $\exp(i\alpha)$ is applied to all ions in the channel conditioned on the bus ion being in the $|1\rangle$-state. This could be achieved by swapping the $|0\rangle$ and $|e\rangle$ population of the bus ion, resonantly driving the $|0\rangle$ population of the channel ion to the excited state and back again with a phase difference of the two fields

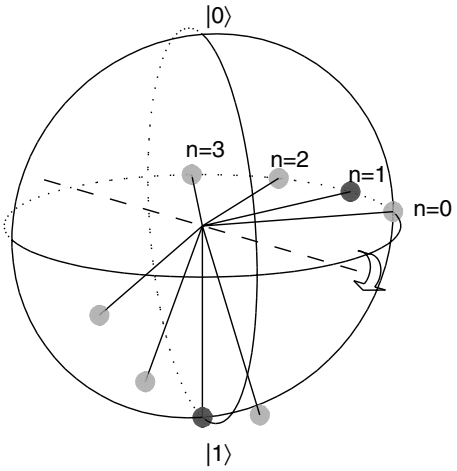

FIG. 3. Graphical illustration of the evolution of the central qubit during the process described for removing superfluous ions.

of α, and finally swapping the bus ion populations back from $|e\rangle$ to $|0\rangle$. Since the resonant process only takes place if the central ion does not shift the transition frequency, i.e., if it is in the $|1\rangle$-state, this will implement the desired conditioned phase shift. The third step is a rotation of the central ion by $\pi/2$ around an axis with the azimuthal angle $\alpha+\pi/2$ on the Bloch sphere: Since the previous step will leave all channel members in their $|0\rangle$ state and the bus ion in the state state $(|0\rangle+\exp(in\alpha)|1\rangle)/\sqrt{2}$, where n is the number of channel members coupled to the ion, this will map the combined state of the central and channel ions to

$$|\psi\rangle = \left(\sin\left(\frac{(n-1)\alpha}{2}\right)|0\rangle + \cos\left(\frac{(n-1)\alpha}{2}\right)|1\rangle\right)|0\rangle^{\otimes n}, \quad (2)$$

up to a global phase, as illustrated in Fig. 3.

The goal of the remaining steps is to stochastically remove some of the channel members if there are more than one: A CNOT on the channel ions conditioned on the state of the central qubit will transfer the channel ions to a state $|\psi\rangle^{\otimes n}$ which has nonvanishing overlap with $|0\rangle^{\otimes n}$ only if $n \neq 1$. We now transfer the channel ions in their $|0\rangle$ state to an excited state (preferably with high branching ratio to $|\text{aux}\rangle$) with a probability amplitude of β and wait for the ions to decay. After this, we perform another CNOT to return all channel ions to their $|0\rangle$ state and re-initialize the bus, after which we repeat the process until all superfluous channel representatives have been removed.

The danger of the above process is that all ions in an instance can decay to their $|\text{aux}\rangle$ state simultaneously. The probability of this happening is β^n, compared to the probability $n\beta^{n-1}(1-\beta)$ of $n-1$ ions decaying, so that by choosing β sufficiently small, we can achieve any desired efficiency at the price of increasing the number of necessary repetitions.

By applying the above procedure repetitively, we can remove superfluous ions and monotonically increase the probability of having precisely one atom per channel.

III. SINGLE INSTANCE QUANTUM COMPUTING WITH A "DESIGNATED READ-OUT ION"

Although the above analysis leaves grounds for some optimism concerning the implementation of quantum computing with not too large registers in the ensemble mode, we believe that full scalability will be more realistically obtained in a single instance implementation. Single ion detection has been demonstrated for Pr and Eu doped crystals [21], but high-fidelity qubit state-selective readout remains difficult because of the >100 μs upper state lifetimes in combination with the absence of a state which can be cycled repetitively as the shelving technique applied in trapped ion studies [22]. To solve this problem we propose to add an additional physical system to the crystal designated only for the readout process. In similarity with the bus ion approach, where the bus ion needs to have strong ion-ion interaction while, e.g., long coherence times is less important, the physical requirements for a readout ion will be different from the qubit ion requirements. For example, as long as the readout ion is able to provide information about the state of the qubit ions, there are no requirements on its coherence times and this opens for a large number of possible readout systems. The readout system could be a molecule, a color center, an ion or some other kind of suitable physical system, but for simplicity we will henceforth refer to the readout system as the "readout ion."

A. Readout ion properties

We will now describe a single instance REQC implementation based on having a designated readout ion. The general idea is to have the readout ion (system) so close to a qubit ion that it can be shifted in and out of resonance with a laser field by transferring the qubit ion between the ground and excited state. In this way the readout system fluorescence signal provides information about the state of the qubit ion.

A reasonable set of requirements on a readout ion (system) could be the following:

(i) The readout system must not be trapped in a nonfluorescent state.

(ii) It should produce a number of output photons/second sufficient for detecting a single readout system.

(iii) It should be possible to have a qubit ion close enough to the readout system to shift it off resonance by an interaction that can be turned on and off.

(iv) The presence of the readout system should not significantly decrease the qubit coherence time.

(v) Readout ion operation should not induce state changes of neighboring qubit ions.

Assume that promoting the qubit ion to the excited state will shift the readout ion into resonance with a laser field at frequency ν_0. To make the discussion more transparent it will be assumed that the qubit ions are Eu ions doped into an Y_2SiO_5 crystal. The excited Eu state that appears best suited for qubit operations has a lifetime of 2 ms. Requiring at least 100 detected photons and assuming a 1% photon detection efficiency, the readout ion should emit at least 1 photon/200 ns. If we further require that the readout process should work in 99% of all cases it must be possible to cycle the readout ion of the order of 10^6 times before it is

trapped in a nonfluorescent state. Regarding the second condition above, the lifetime limited linewidth with a 200 ns excited state is about one MHz. We assume that the readout system has a dipole moment difference $\Delta\mu$, so that one nanometer from a Eu ion, the electric dipole-dipole interaction would induce a frequency shift of $\Delta\mu/(10^{-30}\text{cm GHz})$ (or 3.5 GHz/D). The dipole moment difference for Eu in Y_2SiO_5 is 0.810^{-31}cm (0.023 D). If the readout system has a similarly small dipole moment difference of 0.810^{-31}cm, the line shift one nm away from an Eu qubit ion would be close to 100 MHz.

B. Qubit readout

The system could typically operate with one laser interacting with the qubits (*qubit laser*) and one laser interacting with the readout system (*readout laser*). To readout a qubit (*qubit 1*) close to the readout ion, the readout laser is tuned to frequency ν_0 and a *qubit laser* π pulse is applied to the *qubit 1* $|0\rangle - |e\rangle$ transition. If the readout ion fluoresces, *qubit 1* was in the $|0\rangle$ state.

To readout a neighbor (*qubit 2*) of *qubit 1*, which is not directly coupled to the readout ion, we may either use a quantum gate to swap qubit states 1 and 2, or in a sequential readout of all ions, *qubit 1* can be pumped into the $|0\rangle$ state and a CNOT operation with *qubit 2* as control qubit is carried out to transfer the *qubit 2* state to *qubit 1*. *Qubit 1* can now be readout as described above. It is straightforward to extend the approach to consecutively readout of all qubits in the register.

C. Initialization of single instance computer

We will briefly discuss a design and initiation procedure for the single ion readout scheme. To only readout a single instance it is necessary to have sufficiently low readout ion concentration that there is only one fluorescing readout ion interacting with the laser radiation at frequency ν_0 within the volume observed. Further, before operating the system it has to be fully characterized such that the readout ion frequency and all qubit transition frequencies are known. This is done by first scanning the readout laser across the readout ion inhomogeneous linewidth until readout ion fluorescence is detected. Then the readout laser remains tuned to the readout ion resonance. Now the frequency of the qubit laser is tuned across the inhomogeneous qubit transition and π pulses are applied to the crystal. Once the fluorescence stops, the transition frequency of a qubit (*qubit 1*) that controls the readout ion has been found. By observing the readout ion fluorescence (and applying appropriate operations) the energy level structure of *qubit 1* can now be mapped out. By promoting *qubit 1* to the excited state, the readout ion transition frequency ν_0 when *qubit 1* is excited, can be found. Finding the next qubit, *qubit 2*, is done in a way very similar to finding *qubit 1*. First *qubit 1* is put in the $|0\rangle$ state. The readout laser is tuned to frequency ν_0. Now the frequency of the qubit laser is again tuned across the inhomogeneous qubit transition and π pulses are applied to the crystal. After each π pulse an additional π pulse is applied to the *qubit 1* $|0\rangle - |e\rangle$ transition. This will start the fluorescence signal unless the *qubit 1* transition frequency has been shifted by *qubit 2*. Once the fluorescence does not start after the *qubit 1* $|0\rangle - |e\rangle$ π pulse has been applied, the *qubit 2* frequency is found. The scheme can readily be extended further along the chain using an analogous procedure. The initiation procedure described above requires high efficiency π pulses. However, highly efficient π pulses for these systems have indeed been both designed [10,17] and experimentally demonstrated [7,8,15].

The scheme also has to take into account that there actually will be other ions, spectator ions not part of the qubit chain which, if they become excited, are close enough to interact with ions in the qubit chain. If any of these spectator ions are excited during the operation of the quantum computer they will become entangled with the ions in the chain. Therefore it is necessary to assure that there are no nearby spectator ions in the same frequency channels as those of the ions in the chain. This can be achieved in the following way. Assume there is a spectator ion at the *qubit 2* absorption frequency, ν_2, that spatially is located close enough to interact with ions in the qubit chain. It is assumed that *qubit* 2 can control *qubits 1* and *3* and vice versa. According to Sec. II the probability for having two ions at frequency ν_2 both interacting strongly with *qubit 1* can be made arbitrarily small for low dopant concentration. Exciting *qubit 1* will shift *qubit 2* from its normal absorption frequency, ν_2. However, since none of the other ions absorbing at frequency ν_2 interacts strongly with *qubit 1* their absorption frequency will not shift very much. Optical pumping, following the procedure in Ref. [9], can then be used to remove spectator ions absorbing around frequency ν_2 to other hyperfine states where they are not affected by light pulses at frequency ν_2. Applying this procedure to all qubit frequencies, ν_i, will assure that no spectator ions can interfere with the gate operations.

D. Physical candidate for readout ions

An actual physical readout system needs to be identified, and the $4f-5d$ transition in Ce^{3+} has been proposed as a potentially viable candidate by Guillot-Noël [23]. For Ce^{3+} in YPO_4 the homogeneous linewidth of the $4f-5d$ transition is about 50 MHz and the $5d$ state lifetime is about 20 ns [7]. The corresponding values for the host Y_2SiO_5 could be expected to be similar. Ce^{3+} has a zero nuclear magnetic moment and therefore the ion fluorescence cannot be quenched by decay to a hyperfine state not interacting with the laser at frequency ν_0. The dipole moment difference is unknown but can be expected to be 0.1 D or larger [24,25]. Thus based on these actual and anticipated data, the Ce^{3+} ion appears to be an interesting readout system candidate.

Single-ion readout does not make the system fully scalable in itself. The system increases in size by adding new members to the (possibly branched) chain of qubit ions. Although there are many frequency channels available within the inhomogeneously broadened qubit transition line, a new qubit will eventually happen to have the same frequency as one of the ions already in the chain and these two ions then cannot be individually addressed.

The problem of coinciding transition frequencies can be removed by mounting (closely spaced) electrodes on the

crystal. Eu in Y_2SiO_5 has a Stark coefficient of 35 kHz/(V/cm) [26]. Thus a 1 MV/cm field would shift the transition frequency 35 GHz, a detuning much larger than the inhomogeneous transition linewidth. The part of the qubit chain on which the operations should be carried out can be selected by applying a voltage on the appropriate electrode. All parts of the chain not close to this electrode remain unaffected by light pulses. For such a design the system appears to be fully scalable. Using the ATLAS software (ATLAS Device Simulation Framework from Silvaco International) the electric field distribution has been simulated for the case of a voltage being applied between two 20 nm long electrodes with centers separated by 40 nm, placed on the surface of an insulating material (SiO_2). For a material with a Stark coefficient of 35 kHz/(V/cm), as in Eu doped Y_2SiO_5, these simulations [27] show that shifts of the order of 30 GHz could be obtained within a 60 nm region while ions 20 nm further away were shifted less than 10 GHz.

A more "algorithmic" solution to the problem of degeneracies among the qubit frequencies, is to replace all single qubit operations by two-bit operations, so that operations are applied to "the qubit with frequency ν_i, sitting next to a qubit with frequency ν_j."

IV. CONCLUSION

Experimental work to develop rudimentary precursors of quantum computer hardware in rare-earth-ion-doped crystals have been based on using many instance ensembles of quantum computers in weakly doped crystals. These approaches are, however, not readily scalable to an arbitrary large number of qubits. In this paper scalability requirements for rare-earth-crystal-based quantum computer hardware have been analyzed and in particular two scalable approaches for rare-earth quantum computing have been introduced. One of these is an ensemble-based many instance approach where highly doped crystals (for example stoichiometric crystals [20]) are used to obtain a sufficiently large number of closely situated ions that are able to control each other. The other approach is a single instance scheme where specially designed readout ions are used to readout the values of the qubit ions.

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APPENDIX: ENTANGLING STRENGTH OF THE DIPOLE COUPLING

The dipole gate makes use of the interaction between two ions in their excited states to which they may be selectively promoted from one of the qubit levels. When both ions are excited, the interaction causes an energy shift, which accumulates a phase factor, leading to a nontrivial two-bit gate operation. As suggested in [1,12], it is enough to excite one of the ions and use the shift in resonance frequency to actually avoid excitation of the other one. This implementation is robust against variations of the coupling strength. Since the excited state has a finite lifetime, it is worth looking for proposals that minimize the total time spent by atomic population in the excited states, and one may for example speculate if there is a way to off-resonantly couple both ions, so that the excited state interaction is effective without any of the ions significantly populating that state. We shall now prove that this is unfortunately not possible.

We start by considering the general setting of two quantum subsystems described by Hilbert spaces $\mathcal{H}_a$ and $\mathcal{H}_b$. If the combined system is described by a pure state Ψ in the combined Hilbert space $\mathcal{H}=\mathcal{H}_a\otimes\mathcal{H}_b$, the degree of entanglement between the two systems is quantified by the von Neumann entropy of the reduced density matrix of either one of the systems: $E=S(\rho_a)=S(\rho_b)$.

Calculating E is most easily done if we Schmidt-decompose $|\Psi\rangle$ as follows:

$$|\Psi\rangle = \sum_i c_i |v_i\rangle|u_i\rangle, \tag{A1}$$

where the c_i are real and nonnegative, and $\{|v_i\rangle\}$ and $\{|u_i\rangle\}$ are orthonormal bases of $\mathcal{H}_a$ and $\mathcal{H}_b$, respectively. In terms of the Schmidt decomposition, we have $\rho_a=\Sigma_i c_i^2|v_i\rangle\langle v_i|$, so that $E=S(\rho_a)=-\Sigma_i c_i^2\log_2(c_i^2)$.

To maintain the Schmidt decomposition (3) during evolution, we must allow all of c_i, u_i, and v_i to be time dependent. The time derivative of the Schmidt coefficients is found to be

$$\left.\frac{\partial}{\partial t}c_i^2\right|_{t=0} = \left.\frac{\partial}{\partial t}\langle v_i(0)|\rho_a(t)|v_i(0)\rangle\right|_{t=0}, \tag{A2}$$

since $\rho_a=\Sigma_i c_i^2|v_i\rangle\langle v_i|$ and $\{|v_i(t)\rangle\}$ is an orthonormal basis at all times. To evaluate the derivative, we perform the partial trace over $\mathcal{H}_b$ in the $|u_i(0)\rangle$ basis:

$$\left.\frac{\partial}{\partial t}c_j^2\right|_{t=0} = 2c_j(0)\mathrm{Im}\langle v_j(0)u_j(0)|H|\Psi(0)\rangle, \tag{A3}$$

using the Schrödinger equation and that $\langle v_j u_i|\Psi\rangle=c_j\delta_{i,j}$. Finally, we compute the time derivative of E according to (A4), and using that $\Sigma c_i^2=1$ we find:

$$\dot{E} = 2\sum_{i,j} \log_2\left(\frac{c_j}{c_i}\right) c_i c_j \,\mathrm{Im}(\langle u_i v_i|H|u_j v_j\rangle), \tag{A4}$$

where we note that the diagonal terms do not contribute.

We see that this expression for $\dot{E}$ is additive in H: $\dot{E}(H_a+H_b)=\dot{E}(H_a)+\dot{E}(H_b)$, and that terms of the form $\mathbf{1}\otimes H_a$ corresponding to local operations do not create entanglement: $\dot{E}(\mathbf{1}\otimes H_a)=0$ as we would expect.

According to this argument, the only part of the two-qubit Hamiltonian in REQC capable of producing entanglement is the coupling term $H_c=g|ee\rangle\langle ee|$. The instantaneous rate of creation of entanglement only depends on the instantaneous state of the ions, which, in turn, is controlled by the applied fields.

Using that $a+b\geq 2\sqrt{ab}$ for any nonnegative a and b, we see that

$$w_i \equiv \tfrac{1}{2}(|\langle u_i|e\rangle|^2 + |\langle v_i|e\rangle|^2) \geqslant |\langle u_i v_i|ee\rangle|, \tag{A5}$$

where we have introduced $\{w_i\}$, which are seen to fulfill $\Sigma_i w_i = 1$ as $\{|u_i\rangle\}$ and $\{|u_i\rangle\}$ are orthonormal bases. Inserting H_c into the expression (A4), the inequality (A5) allows us to establish the following bound on the rate of entanglement creation:

$$\frac{|\dot{E}|}{g} \leqslant 2\sum_i c_i^2 w_i \sum_j f(\theta_{i,j}) w_j,$$

where we have parametrized (c_i, c_j) as $(\sin(\theta_{i,j}), \cos(\theta_{i,j}))(c_i^2 + c_j^2)$, and introduced $f(\theta) = |\log(\tan(\theta))|\sin(\theta)\cos(\theta)$. A numerical analysis shows that f is bounded by 0.478…, and hence

$$|\dot{E}| < \frac{1}{2} P_e^{(\mathrm{tot})} g, \tag{A6}$$

where $P_e^{(\mathrm{tot})} = \langle\Psi|(\mathbf{1}\otimes|e\rangle\langle e| + |e\rangle\langle e|\otimes\mathbf{1})|\Psi\rangle = 2\Sigma_i w_i c_i^2$ is the total excited state population.

The implications of Eq. (A6) are clear: in order to be able to obtain one unit of entanglement, as required for a universal two-qubit gate operation, the system must necessarily suffer an integrated excitation $\int P_e^{(\mathrm{tot})} dt > 2/g$, where g is the dipole coupling strength.

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