

## Quantum computation with rotational states of nonpolar ionic molecules

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(Received 20 October 2012; revised manuscript received 13 January 2013; published 4 April 2013)

We propose a quantum computer architecture that is robust against decoherence and scalable. As a qubit we adopt rotational states of a nonpolar ionic molecule trapped in an ion trap. It is revealed that the rotational-state qubits are much more immune to decoherence than the conventional electronic-state qubits of atomic ions. A complete method set that includes state preparation, a single-qubit gate, a controlled-NOT gate, and qubit readout suitable for the rotational-state qubits is provided. Since the ionic molecules can be transported in an array of ion traps, the rotational-state qubits are expected to be a promising candidate to build a large-scale quantum computer.

DOI: [10.1103/PhysRevA.87.040302](https://doi.org/10.1103/PhysRevA.87.040302)

PACS number(s): 03.67.Lx, 33.20.Sn, 37.10.Ty, 42.50.Ex

A quantum computer (QC) is a computing machine that uses quantum logic totally different from the Boolean logic on which classical computers are built. If a QC is successfully built, it will make numerous unimaginable things possible, in a similar manner that lasers have done [1]. The most critical difficulty in building a QC is that qubits (two-level quantum systems) should be well isolated from environment to preserve their fragile quantum states from decoherence, while at the same time a quantum channel should exist to give programmers access to the qubits. Another serious difficulty, called scalability, arises from the fact that one should manipulate at least thousands of qubits to perform a practical computation [2]. Since all elementary building blocks necessary for construction of a QC have been demonstrated over the past decade, current goals intensively pursued are scaling up to a larger number of qubits and raising the fidelity of qubit manipulation [3]. In accomplishing these missions, the most critical obstacle is the decoherence problem. Thus it is desirable to find an alternative qubit system immune to decoherence.

We propose an ion-trap-based QC architecture that is robust against decoherence and scalable. As a qubit we adopt rotational states of a nonpolar molecular ion trapped in an ion trap; rotational states of neutral polar molecules have already been proposed as qubits [4,5]. Without collisions, the lifetime of rotational states of a nonpolar molecule can reach several years [6] because the rotational state is electromagnetically inactive owing to no permanent dipole moment. We reveal that the rotational-state qubits of nonpolar molecules are much more immune to decoherence than the conventional electronic-state qubits of atomic ions and provide a complete method set necessary for building a QC that consists of state preparation, a single-qubit gate, a controlled-NOT gate, and qubit readout appropriate to the rotational-state qubits. Since we adopt ionic molecules, one can utilize well-developed ion-trap technologies such as separation, transportation, and combination of individual ions, which are the key ingredients to implementing a large-scale QC [7]. The movement control has already been demonstrated for atomic ions [8] and no

additional restriction is imposed on molecular ions. Because nonpolar quantum rotors are excellent gyroscopes, their rotational states should be very robust under the transportation.

We first survey general features of rotational states of nonpolar linear molecules and select two levels as a qubit. Rotationally stationary states of a linear molecule are angular momentum eigenstates  $|J, M\rangle$ , where  $J$  is the angular momentum quantum number and  $M$  is its  $z$ -axis projection in the laboratory frame. Because a nonpolar molecule has no permanent dipole moment, the laser field cannot cause dipole transitions ( $\Delta J = \pm 1$ ). However, since linear molecules have anisotropy in their polarizability, they allow rotational two-photon Raman transitions with  $\Delta J = \pm 2$ . When the laser field is linearly polarized along the  $z$  axis, one additional selection rule gives  $\Delta M = 0$ . With these selection rules, one can choose a qubit system as  $|\downarrow\rangle \equiv |J_0, M_0\rangle$  and  $|\uparrow\rangle \equiv |J_0 + 2, M_0\rangle$  for arbitrary  $J_0$  and  $M_0$ . However, the choice of  $J_0$  and  $M_0$  is restricted according to molecular species, because nuclear spin statistics categorize nonpolar linear molecules into three groups: even-, odd-, and all- $J$  molecules [9]. In order to be a nonpolar molecule, the nuclei at both ends of the molecule should be identical particles obeying the exchange symmetry of quantum mechanics. For example, in  $\text{NS}_2^+$ , the two  $^{32}\text{S}$  are spin-zero bosons and the electronic ground state is  $^1\Sigma_g^+$  so that only even- $J$  states are allowed. Among the three types, the even- $J$  species is the most preferred because the rotational ground state of this kind is  $|0, 0\rangle$ , which is nondegenerate. In contrast, the ground state of an odd- $J$  molecule is degenerate and can be  $|1, -1\rangle$ ,  $|1, 0\rangle$ , or  $|1, 1\rangle$  so that an additional selection is needed to prepare a qubit state even at zero temperature. Moreover, the ground state of an all- $J$  molecule is more ambiguous because it depends on the spin states of the nuclei. In this Rapid Communication we assume that an even- $J$  molecule is chosen and the qubit system is  $|\downarrow\rangle \equiv |0, 0\rangle$  and  $|\uparrow\rangle \equiv |2, 0\rangle$ . Because the Hilbert space of rotational states of an even- $J$  molecule intrinsically excludes odd- $J$  states, one additional merit arises that there is no possibility of state leakage into unwanted odd- $J$  space.

In terms of decoherence, we compare the rotational-state qubits with conventional electronic-state qubits. In an ion-trap QC, the major source of decoherence is a fluctuating magnetic field [10]. In the conventional atomic-ion architecture, qubits

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are made of electronic states of alkaline-earth-metal ions such as  $\text{Be}^+$  or  $\text{Mg}^+$ , which have only one open-shell electron. This makes electronic spin states a doublet with a big magnetic dipole moment, which results in a strong coupling between the qubit and the magnetic field through the Zeeman effect. In contrast, in the case of the molecular rotational-state qubits, the magnetic moment of electrons affects the qubit state merely indirectly. Furthermore, there exist molecular ions having no electronic magnetic moment such that the electronic ground state is  $^1\Sigma$  state. In that case, the magnetic field affects the rotational-state qubit only through the rotational magnetic moment  $\mu_r$ , which is proportional to the rotational  $g$  factor  $g_r$  and the rotational angular momentum  $J$  as expressed by  $\mu_r = g_r \mu_N \sqrt{J(J+1)}$  [9], where  $\mu_N$  is the nuclear magneton. Most nonpolar molecules have  $g$  factors smaller than 1 [9], so  $\mu_r$  is very small compared to the electronic spin magnetic moment, which is equal to one Bohr magneton  $1840\mu_N$ , which the conventional electronic-state qubits have. It is thus expected that the rotational-state qubits are very robust against decoherence from magnetic field fluctuations.

Several techniques have been adopted to improve the coherence time of atomic ions. For example, one can choose electronic-state qubits that are insensitive under magnetic fields to first order [10,11] and can adopt the decoherence-free subspace (DFS) technique [12,13]. With these techniques, the single-qubit coherence time of an atomic ion has been achieved to be about 20 s [13]. However, 20 s is not sufficient for a large-scale quantum computer because when the coherence time of a single qubit is  $\tau$ , that of  $N$  qubits is approximately  $\tau/N$  [14]. The limited coherence time is mainly due to a high-order Zeeman shift and local field fluctuation different at each ion composing a DFS pair. These two effects will be reduced if the magnetic moment of a qubit is small. Thus the tiny magnetic moment of the rotational-state qubit can greatly extend the coherence time several orders longer than the conventional electronic-state qubits of atomic ions. Furthermore, the rotational-state qubit is also first-order insensitive to the magnetic field because we choose the qubit states with  $M = 0$  and the DFS technique can also be applied to rotational-state qubits.

We examined a variety of molecular ions to choose a suitable candidate that satisfies the requirements including linearity, nonpolarity, even- $J$  state, electronic  $^1\Sigma$  state, and small rotational  $g$  factor. If we choose the candidate among singly charged molecular ions, triatomic molecular ions, such as  $\text{NO}_2^+$  and  $\text{NS}_2^+$ , can fulfill the requirements. Considering rotational  $g$  factors calculated by the DALTON software [15], we suggest  $\text{NS}_2^+$  as a suitable candidate due to its tiny rotational  $g$  factor of  $-0.014$ , which means that the magnetic moment of  $|\downarrow\rangle$  is zero due to  $J = 0$  and that of  $|\uparrow\rangle$  is  $-0.034\mu_N$ . If we extend our choice to doubly charged molecular ions, homonuclear diatomic dications can also be chosen. Although most diatomic dications are metastable, the lifetimes of some species are very long; e.g.,  $\text{Ba}_2^{2+}$  has a lifetime longer than the age of the universe [16]. The diatomic dication  $\text{Ba}_2^{2+}$  not only fulfills the above requirements, but also has the advantage of no nuclear spin. Because the nuclear spin causes the hyperfine spin-rotation coupling [17], the nuclear spin can be another source of decoherence. While singly charged triatomic  $^1\Sigma$  molecular ions cannot have zero nuclear spin in their center

nucleus, diatomic dications can simultaneously fulfill the two requirements of a  $^1\Sigma$  electronic state and zero nuclear spin. In this respect, very-long-lived homonuclear diatomic dications such as  $\text{Ba}_2^{2+}$  can also be proper candidates.

The first step in operating a QC is the state preparation of qubits, which should include translational, vibrational, and rotational cooling of molecular ions. The translational cooling of molecular ions can be achieved by the sympathetic cooling method [18], which relies on cotrapped atomic alkaline-earth-metal ions that can be easily cooled down using a laser. Sophisticated techniques are needed for cooling of rovibrational states of molecules after the translational cooling and experimental demonstrations were reported only recently and moreover only for polar molecular ions [19,20]. For nonpolar molecular ions, a method that exploits the motional ground state shared with atomic cooling-pair ions was proposed [21,22]. Although its experimental demonstration has not been reported yet, this method is expected to make nonpolar molecular ions reach the rotational ground state, i.e.,  $|\downarrow\rangle$ , with high fidelity of more than 99.9% [22].

A single-qubit operation can be performed through a resonant two-photon Raman transition between qubit levels. The Hamiltonian for rotational states of a nonpolar molecule interacting in a laser field is given by [23,24]

$$\mathbf{H}(t) = B_0 \mathbf{J}^2 - \frac{1}{2} \Delta\alpha \mathcal{E}^2(t) \cos^2\theta, \quad (1)$$

where  $B_0$  is the molecular rotational constant,  $\Delta\alpha \equiv \alpha_{\parallel} - \alpha_{\perp}$  is the anisotropy of molecular polarizability,  $\mathcal{E}(t)$  is the laser electric field, and  $\theta$  is the angle between the laser polarization and molecular axis. Because the energy of  $|J, M\rangle$  is  $B_0 J(J+1)$ , the energy gap between qubit levels is  $\omega_0 = 6B_0$ . A pair of laser beams linearly polarized along the  $z$  direction that have frequencies  $\omega_1$  and  $\omega_2$  with the frequency difference equal to  $\omega_0$  can cause the resonant two-photon transition to achieve the single-qubit operation, as shown in Fig. 1. Here the laser beams are preferably IR lasers detuned far from vibrational or electronic transitions of the molecular ion. If the amplitudes of the two laser beams are equally  $\mathcal{E}_0/2$ , the synthesized field

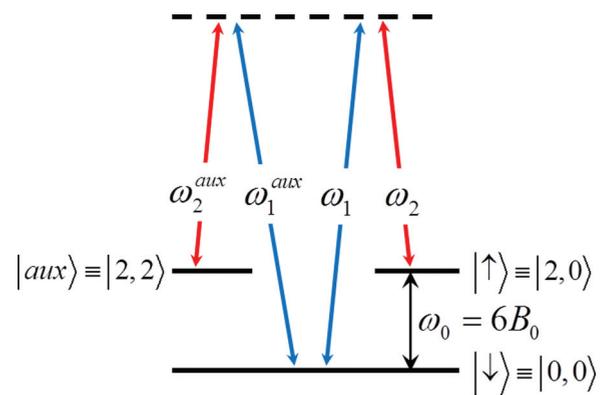


FIG. 1. (Color online) Energy-level diagram of qubit states. A pair of linearly polarized laser beams with frequencies  $\omega_1$  and  $\omega_2$  induces a resonant two-photon Raman transition between the qubit levels for a single-qubit gate. The state  $|\text{aux}\rangle$  and two circularly polarized lasers with frequencies  $\omega_1^{\text{aux}}$  and  $\omega_2^{\text{aux}}$  are used when the Cirac-Zoller controlled-NOT gate is adopted.

is given by

$$\vec{\mathcal{E}}_{|\downarrow\rangle\leftrightarrow|\uparrow\rangle}(t) = \hat{\mathbf{z}}\mathcal{E}_0 \cos \frac{\omega_1 + \omega_2}{2} t \cos \left( \frac{\omega_0}{2} t + \varphi \right) \quad (2)$$

and brings about the resonant Raman-Rabi oscillation with the Rabi frequency

$$\Omega = \frac{1}{8\hbar} \Delta\alpha \mathcal{E}_0^2 \langle \downarrow | \cos^2 \theta | \uparrow \rangle = \frac{0.2981}{8\hbar} \Delta\alpha \mathcal{E}_0^2. \quad (3)$$

To be specific, the properties of  $\text{NS}_2^+$  were calculated using the GAUSSIAN software [25] to obtain  $B_0 = 3.44$  GHz, leading to  $\omega_0 = 20.64$  GHz, and  $\Delta\alpha = 8.47 \text{ \AA}^3$ . In order to achieve a Rabi frequency  $\Omega$  of, e.g., 1 MHz with this molecule, a field intensity (corresponding to  $\mathcal{E}_0^2$ ) of  $2.5 \times 10^6 \text{ W/cm}^2$  is needed. It is desirable that the two laser beams are copropagating in order not to change the motional state during the single-qubit operation since the photon recoil force exerted on ions becomes minimal in that condition [26].

The intermediate state in Fig. 1 is not a detuned real physical state but a virtual state because the interaction Hamiltonian of Eq. (1) induces the selection rule  $\Delta J = \pm 2$ , which intrinsically requires two photons. This eliminates worry about detuning and state leakage into the intermediate state, which conventional electronic-hyperfine qubits of atomic ions suffer from. In this respect, the rotational-state qubits are expected to achieve a high fidelity and a high speed in gate operations owing to the lack of state leakage into the unwanted intermediate state. This is another advantage of the rotational-state qubits.

In every proposed QC architecture, a key issue has been how a controlled-NOT gate is realized. Since any quantum logic gate can be decomposed into single-qubit operations and two-qubit controlled-NOT operations, the two kinds of operations constitute universal quantum gates [27]. A big advantage of the ion-trap-based QC is that the ion trap provides an efficient way to carry out the controlled-NOT gate via collective motional states that the ions share. With the aid of the motional state, qubits can be effectively coupled even if their direct interaction is very weak. Several proposals have been made to realize the controlled-NOT gate in an ion trap [2,28–30] and some of them were demonstrated with electronic-state qubits of atomic ions [31–33]. Most of these proposals can also be applied to the rotational-state qubits of molecular ions as long as the ions are cotrapped in a trap potential.

We consider two representative proposals and provide concrete methods to implement the proposals with the rotational-state qubits. Let us first consider the controlled-NOT gate proposed by Cirac and Zoller [28]. The Cirac-Zoller gate needs an auxiliary state  $|\text{aux}\rangle$  coupled with  $|\downarrow\rangle$  by another laser without affecting  $|\uparrow\rangle$ . For the rotational-state qubit, we suggest  $|\text{aux}\rangle \equiv |2,2\rangle$ , as shown in Fig. 1. The transition between  $|\downarrow\rangle$  and  $|\text{aux}\rangle$  can be performed by two circularly polarized laser beams whose polarization directions are counter-rotating in the  $x$ - $y$  plane. If the two beams have frequencies  $\omega_1^{\text{aux}}$  and  $\omega_2^{\text{aux}}$  with frequency difference  $\omega_0$  and their amplitudes are equally  $\mathcal{E}_0/2$ , the synthesized field is formed as

$$\vec{\mathcal{E}}_{|\downarrow\rangle\leftrightarrow|\text{aux}\rangle}(t) = \mathcal{E}_0 \cos \frac{\omega_1^{\text{aux}} + \omega_2^{\text{aux}}}{2} t \left\{ \hat{\mathbf{x}} \cos \left( \frac{\omega_0}{2} t + \varphi \right) + \hat{\mathbf{y}} \sin \left( \frac{\omega_0}{2} t + \varphi \right) \right\}. \quad (4)$$

This is a linearly polarized field whose polarization direction is rotating in the  $x$ - $y$  plane with a frequency of  $\omega_0/2$ , resulting in the selection rules  $\Delta J = \pm 2$  and  $\Delta M = \pm 2$ . Owing to the selection rules,  $\vec{\mathcal{E}}_{|\downarrow\rangle\leftrightarrow|\text{aux}\rangle}$  causes the transition between  $|\downarrow\rangle$  and  $|\text{aux}\rangle$  without affecting  $|\uparrow\rangle$ . This exclusive transition is sufficient to implement the Cirac-Zoller gate. In contrast to the single-qubit gate, the Cirac-Zoller gate should generate the transition not only between  $|\downarrow\rangle$  and  $|\text{aux}\rangle$  but also between the collective motional states  $|n=1\rangle$  and  $|n=0\rangle$ , where  $n$  is the phonon quantum number. In order to alter the motional state, the actual frequency difference of the two laser beams is red detuned such that  $\omega_0 \rightarrow \omega_0 - \nu$ , where  $\nu$  is the phonon mode frequency along the  $z$  axis. In addition to this detuning, if the laser beams are counterpropagating, the motional state is more efficiently changed than in the copropagating condition [26]. Additional requirements of this gate are that the motional state has to be cooled down to zero temperature and the laser beams should be addressed to one particular ion.

These requirements were released in another controlled-NOT scheme proposed by Sørensen and Mølmer [29]. The Sørensen-Mølmer gate is performed through off-resonance laser pulses illuminating two ions simultaneously and does not need the auxiliary state. Because this method uses the collective motional state as a virtual intermediate state, the two-qubit gate works even in a motional thermal state. There is no restriction in adopting this method for the rotational-state qubits since the method works also in Raman transitions [29].

At the end of computation, final qubit states should be read out. Previously, the state-of-the-art method of qubit readout has been based on fluorescence detection [34]. Unfortunately, the fluorescence-based method cannot be directly applied to the rotational-state qubits because in the rotational states there is no cycling transition decaying back to one of the qubit states. However, in an ion trap where two ions are cotrapped, an internal state of one ion can be transferred to that of the other ion [35]. With this technique, the rotational-state qubit of a molecular ion can be read out by instead measuring the electronic-state qubit of an atomic ion that has a good cycling transition. Figure 2 illustrates this readout scheme. Alkaline-earth-metal ions such as  $^9\text{Be}^+$  and  $^{25}\text{Mg}^+$  have a good cycling transition and their hyperfine qubits can be measured with high fidelity. Among them, one having a mass similar to that of the molecular ion is preferred because the motional interaction between the two ions can be efficient [21]. If one chooses the  $\text{NS}_2^+$  molecule, we recommend  $^{87}\text{Sr}^+$  due to their similar mass. The readout scheme consists of five steps. (i) The shared motional state is sympathetically cooled down to zero temperature by laser cooling of the atomic ion. (ii) The atomic ion undergoes state preparation to  $|\downarrow_{\text{atom}}\rangle$ . (iii) A blue-sideband Raman transition from  $|\uparrow\rangle$  to  $|\text{read}\rangle$  takes place by applying a  $\pi$  pulse detuned by  $\nu$ , where  $\nu$  is the motional mode frequency, which means  $\omega_1^{\text{read}} - \omega_2^{\text{read}} = \omega_0^{\text{read}} + \nu$ . In this step, the motional state is changed from  $|n=0\rangle$  to  $|n=1\rangle$  only if the rotational-state qubit is  $|\uparrow\rangle$ . It is preferable for the two Raman beams to counterpropagate along the motional axis in order to efficiently change the motional quantum number [26]. (iv) A red-sideband Raman transition of the atomic ion from  $|\downarrow_{\text{atom}}\rangle$  to  $|\uparrow_{\text{atom}}\rangle$  occurs. Only when the motional state is excited to  $|n=1\rangle$  does this red-sideband

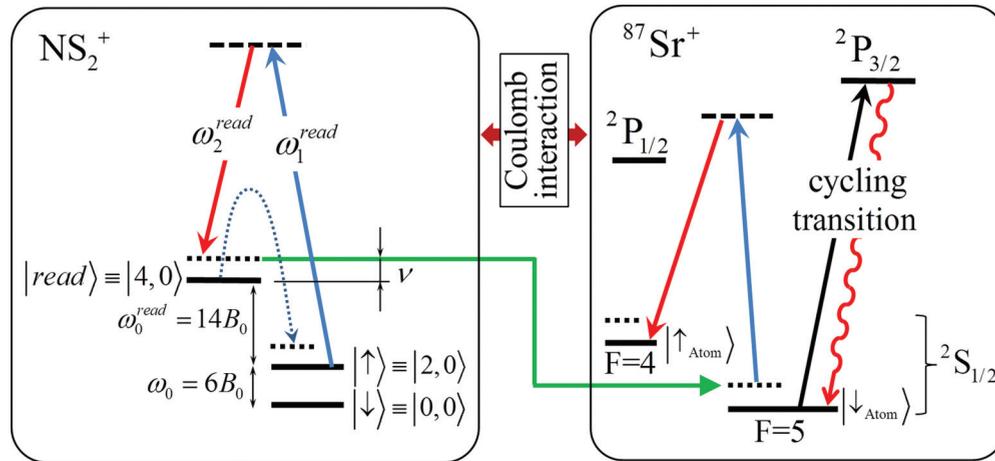


FIG. 2. (Color online) Qubit readout scheme using the state-transfer technique. Rotational-state qubits of molecular ions are read out by instead measuring electronic-state qubits of cotrapped atomic ions with fluorescence detection. Energy levels with dotted lines indicate the motional excited state  $|n = 1\rangle$  with corresponding internal states.

transition actually occur. (v) The state-dependent fluorescence of the atomic ion is detected using the cycling transition. If fluorescence is detected, the atomic state is measured as  $|\downarrow_{\text{atom}}\rangle$  and the rotational-state qubit is considered to be measured as  $|\downarrow\rangle$ ; otherwise it is  $|\uparrow\rangle$ .

In order to enhance the fidelity of readout, the above procedures can be repetitively performed, with slight modification of step (iii). Because at the second repetition the rotational state would lie on  $|\text{read}\rangle$  (not on  $|\uparrow\rangle$ ) with a motional ground state  $|n = 0\rangle$ , the red-sideband Raman transition from  $|\text{read}\rangle$  to  $|\uparrow\rangle$  (see the dotted arrow in Fig. 2) will excite the motional state. This causes the same effect as the former procedures and can transfer quantum states to the atomic ion. This way, readout can be performed several times. In every step (iii), the blue- and red-sideband transitions alternate. This repetitive readout scheme is almost the same as the experiment done by Hume *et al.* [35]. The only difference is that, in our case, the lifetime of  $|\text{read}\rangle$  is very long (years), so  $|\text{read}\rangle$  does not decay back to  $|\uparrow\rangle$ . This long lifetime of rotational states of nonpolar molecules is rather advantageous in achieving high fidelity. In

the experiment of Hume *et al.*, there was about 1% probability of state leakage during the decay process, limiting the fidelity. In our case, there is no concern about state leakage owing to no decay. In this respect, our proposed readout scheme is expected to achieve at least 99.94% fidelity, which Hume *et al.* already achieved.

In conclusion, we have proposed a QC architecture using rotational-state qubits of nonpolar ionic molecules. It was shown that the rotational-state qubits are very immune to decoherence and can achieve high-fidelity logic gates owing to no state leakage into an intermediate state during qubit manipulations. A complete method set necessary for building a QC suitable for the rotational-state qubits was provided. The proposed architecture is expected to be one of the promising candidates for implementing a practical QC.

We thank Hai Woong Lee, Jaewan Kim, and Jaewook Ahn for useful comments. This work was supported by the Ministry of Education, Science and Technology of Korea through the National Research Foundation and Institute for Basic Science.

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