

Measurements on trapped laser-cooled ions using quantum computations

C. D'Helon* and G. J. Milburn

Department of Physics, University of Queensland, St. Lucia 4072, Australia

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We show that a series of quantum computations involving an isolated N -quantum-bit ion register can be used to make an approximate quantum nondemolition measurement of the number state distribution of a collective vibrational mode. A unitary transformation is applied to the Fourier transformed state of the ion register to copy the vibrational statistics to the electronic mode, and the number state distribution is then measured in the electronic number state basis using the quantum jumps technique. [S1050-2947(96)04912-8]

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I. INTRODUCTION

Laser-cooled ions confined in a linear rf trap, and interacting with optical standing waves are a feasible implementation of a quantum computer. In such a system quantum bits (qubits) are represented by internal ion transitions, and quantum logic gates can be realized by coupling ions through a collective vibrational mode, as proposed by Cirac and Zoller [1]. The relatively large decoherence time of ion traps is crucial in enabling long sequences of coherent quantum computations to be performed. Also, the value of qubits can be determined very efficiently by monitoring quantum jumps using probe resonance fluorescence measurements [2,3].

In a recent experiment, Monroe *et al.* [4] have demonstrated the operation of a two-qubit universal quantum logic gate using a single trapped laser-cooled ion. One qubit consisted of two hyperfine electronic states and the other of the two lowest vibrational levels of the ion. The ratio of the decoherence rate of the trap to the switching rate of the universal gate was favorable for the purposes of simple quantum computations, and an extension of the system to more qubits looks promising.

For a single ion trapped in a harmonic potential, the initial vibrational population distribution can be determined from measurements of the population inversion, as discussed by Cirac *et al.* [5]. Several measurement schemes have also been proposed to determine the initial vibrational density matrix of a single ion trapped in a harmonic potential, employing tomographical reconstructions of phase-space distributions of the ion [6–8].

In this paper, we show that quantum computations on an N -qubit ion register can be used to measure the number state distribution of a vibrational mode common to the ions. This is a specific example of a technique that may have wider implications for quantum limited measurements. By making quantum computational transformations on the output state of an apparatus prior to a final readout, a much wider class of operators can be measured. In simple terms, instead of measuring a difficult operator directly on the output state, a quantum computer can be used to unitarily transform the output state so that measurement of a simple operator would yield the same statistics.

Starting with all the ions in the ground state, we apply a Fourier transform to the initial state using single-qubit rotations. A set of far-detuned optical standing wave pulses are then turned on to entangle the electronic and vibrational number states of the ion register. Finally, an inverse Fourier transform is applied to the system. The resultant state displays a 2^N -periodic correlation between vibrational number states and corresponding electronic number states.

For a sufficiently large number of ions, this scheme is a quantum nondemolition (QND) measurement of the initial vibrational number state distribution. The initial occupation number of the n th harmonic oscillator state is given by measuring the relative frequency of the electronic number state $|n\rangle_e$, which encodes the electronic states of the ions arranged in a definite order.

Adopting a different point of view, if the initial vibrational number state distribution is known, our measurement scheme can be used to create entangled ion states in an alternative manner to that discussed by Cirac and Zoller [1].

II. MEASUREMENT MODEL

Our physical system consists of N ions forming a crystalized string confined in a linear rf trap, and our measurement scheme aims to determine the number state distribution of a vibrational mode that is common to all the ions. Each ion is positioned within a different strongly focused optical standing wave, which interacts with the two-level atomic transition chosen to implement a qubit. The spontaneous decay rate from the upper levels of qubits is assumed to be negligible on the time scales of interest; i.e., the upper levels are metastable, in order to avoid decoherence effects. The ions experience harmonic motion about their equilibrium positions at the trap frequency ν , and are laser cooled into the Lamb-Dicke limit so that their vibrational amplitudes are much less than the wavelength of the light. The laser cooling process restricts the motion of the ions to the lowest vibrational mode along the axis that the ions are trapped on, and realizes the common vibrational mode we are interested in.

The electronic basis states $|k\rangle_e$ used for the ion register are defined by the product of the individual electronic eigenstates of the qubits arranged in some definite order,

$$|k\rangle_e = |S_N\rangle_N \otimes |S_{N-1}\rangle_{N-1} \otimes \cdots \otimes |S_1\rangle_1, \quad (1)$$

*Electronic address: dhelon@wilson.physics.uq.oz.au

where $S_i=0,1$ represents a ground or excited state, respectively. These electronic number states are labeled by the integer k ,

$$k = S_N \times 2^{N-1} + S_{N-1} \times 2^{N-2} + \dots + S_1 \times 2^0, \quad (2)$$

encoding the binary string $S_N S_{N-1} \dots S_1$, ($0 \leq k \leq 2^N - 1$).

We consider a Hamiltonian H , which is obtained by applying a set of standing wave pulses to the ion register, with the equilibrium position of each ion placed at a node of the corresponding standing wave. This Hamiltonian couples the vibrational and electronic modes, and is given by

$$H = \hbar \sum_{j=1}^N \Delta_j \sigma_z^{(j)} + \hbar \nu a^\dagger a + \frac{\hbar}{2\sqrt{N}} \times \sum_{j=1}^N \Omega_j (\sigma_+^{(j)} + \sigma_-^{(j)}) \sin[\eta_j (a + a^\dagger)], \quad (3)$$

where a, a^\dagger are the annihilation and creation operators of the vibrational mode, and $\sigma_z^{(j)}, \sigma_+^{(j)}, \sigma_-^{(j)}$ are the population inversion, raising, and lowering operators of the j th qubit, respectively. The Rabi frequencies Ω_j and the detuning frequencies Δ_j between the atomic and standing wave frequencies ($\Delta_j = \omega_0^{(j)} - \omega_L^{(j)}$) are associated with the j th qubit. If we scale the position of each ion in terms of the rms position fluctuations around its equilibrium point, the dimensionless Lamb-Dicke parameters are given by $\eta_j = K_j \sqrt{\hbar/2m_j\nu}$, where K_j is the wave number of the standing wave interacting with the j th qubit, and m_j is the mass of the j th ion.

The detuning frequencies are set to the same frequency, i.e., $\Delta_j = \Delta$, which is assumed to be much greater than the trap frequency ($|\Delta| \gg \nu$) and well away from resonant vibrational frequencies. We show in the Appendix that when the standing waves are turned on in this far-detuned regime, with the ions in the Lamb-Dicke limit ($\eta_j \ll 1$), the above Hamiltonian can be expressed in an interaction picture as

$$H'_I = \hbar a^\dagger a \sum_{j=1}^N \chi_j (\sigma_z^{(j)} + \frac{1}{2}) \quad (4)$$

for interaction times $t \gg \nu^{-1}$, where $\chi_j = \eta_j^2 \Omega_j^2 / (N\Delta)$.

The Hamiltonian H'_I generates a unitary transformation

$$U = \exp\left(\frac{-i2\pi a^\dagger a Y}{2^N}\right) \quad (5)$$

if we adjust the product of the parameters χ_j and the durations τ_j of the standing wave pulses to increase geometrically with qubit number, i.e., $\chi_j \tau_j = 2^j \pi / 2^N$. The electronic operator Y provides a binary ordering of the qubits,

$$Y = \sum_{j=1}^N (\sigma_z^{(j)} + \frac{1}{2}) 2^{j-1}, \quad (6)$$

and is an eigenoperator of the electronic number states

$$Y = \sum_{k=0}^{2^N-1} k |k\rangle_e \langle k|. \quad (7)$$

For a single ion, the transformation U accompanied by simple qubit rotations is equivalent to the unitary transformation used by Monroe *et al.* [4] to implement a quantum controlled-not gate.

In the case of an N -ion register, the transformation U does not affect the vibrational state, however it displaces the eigenstates of an electronic operator Φ , which is canonically conjugate to Y . The eigenstates of Φ are a complementary set of electronic basis states $|\tilde{p}\rangle_e$ given by Fourier transforms of the electronic number states $|k\rangle_e$,

$$|\tilde{p}\rangle_e = \frac{1}{\sqrt{2^N}} \sum_{k=0}^{2^N-1} \exp\left(\frac{-2\pi i k p}{2^N}\right) |k\rangle_e \quad (8)$$

for $0 \leq p \leq 2^N - 1$. The transformation U displaces these eigenstates by the vibrational number,

$$U |n\rangle_{\text{vib}} \otimes |\tilde{p}\rangle_e = |n\rangle_{\text{vib}} \otimes |\tilde{\mathcal{N}}_p\rangle_e, \quad (9)$$

where $|n\rangle_{\text{vib}}$ is a vibrational number state, and $\tilde{\mathcal{N}}_p = (p + n) \pmod{2^N}$. Hence we can displace the Fourier-transformed zero state $|\tilde{0}\rangle_e$ in order to copy the statistics of the vibrational distribution to the electronic mode.

It would be ideal to measure the operator Φ after the transformation U , to determine the vibrational statistics directly from the resultant state; however, it is unclear how to measure this observable on the ion register. On the other hand, measurements of Y are readily feasible using the quantum jumps technique, hence we unitarily transform the ion register after interaction in such a way that a measurement of Y on the transformed output state is equivalent to measuring Φ on the untransformed output state.

The initial vibrational state of the ion register is arbitrary, and we assume that all the ions start in the electronic ground state, so that the initial state of the ions is given by

$$|\psi_i\rangle = \sum_{n=0}^{\infty} c_n |n\rangle_{\text{vib}} \otimes |0\rangle_e. \quad (10)$$

The ion register is then prepared in a superposition of all possible electronic number states by rotating each qubit. The rotations are accomplished by placing the equilibrium positions of the ions to coincide with antinodes of the respective standing waves, and applying a set of resonant $\pi/2$ pulses (with $-\pi/2$ laser phase) to set each qubit to a superposition of the ground and excited states. This transformation does not affect the vibrational state, and is equivalent to applying a Fourier transform to the initial state, such that $|0\rangle_e \rightarrow |\tilde{0}\rangle_e$.

The equilibrium positions of the ions are then placed to coincide with nodes of the respective standing waves, and a series of pulses are applied in the far-detuned regime. The detuning frequency Δ is well away from vibrational resonant frequencies, and the ions are in the Lamb-Dicke limit so that the interaction is well described by the Hamiltonian H'_I from Eq. (4), which was derived in the Appendix. The relative intensities and durations of the standing wave pulses can be adjusted to achieve the geometric variation of $\chi_j \tau_j$ required to obtain the transformation U given in Eq. (5). The resultant

state of the ion register $|\psi_u\rangle$ is a sum of displaced electronic Fourier-transformed states entangled with corresponding vibrational number states,

$$|\psi_u\rangle = \sum_{n=0}^{\infty} c_n |n\rangle_{\text{vib}} \otimes |\tilde{\mathcal{N}}_0\rangle_e, \quad (11)$$

where $\mathcal{N}_0 = n \pmod{2^N}$, and can be rewritten as

$$|\psi_u\rangle = \sum_{n=0}^{\infty} \sum_{p=0}^{2^N-1} c_{p+n2^N} |p+n2^N\rangle_{\text{vib}} \otimes |\tilde{p}\rangle_e. \quad (12)$$

Since it is difficult to measure the observable Φ , an inverse Fourier transform is applied to the ion register to express the final state $|\psi_{\text{out}}\rangle$ in terms of the electronic basis states $|k\rangle_e$,

$$|\psi_{\text{out}}\rangle = \sum_{n=0}^{\infty} \sum_{k=0}^{2^N-1} c_{k+n2^N} |k+n2^N\rangle_{\text{vib}} \otimes |k\rangle_e. \quad (13)$$

This is achieved formally via the transformation $|\tilde{p}\rangle_e \rightarrow |p\rangle_e$, which is equivalent to a Fourier transformation $|k\rangle_e \rightarrow \sum_{l=0}^{2^N-1} e^{2\pi i l k / 2^N} |l\rangle_e$ of each electronic number state. The experimental implementation of this unitary transformation involves a series of one-bit and two-bit quantum gates, using a scheme such as that devised by Coppersmith [9,10].

The one-bit quantum gates correspond to simple qubit rotations [1], and are relatively straightforward to implement in the same way we set up the initial electronic state of our ion register to a superposition of all number states. A resonant $\pi/2$ standing wave pulse (with $-\pi/2$ laser phase) is applied to the appropriate qubit to rotate its levels, with the equilibrium position of the ion placed at an antinode of the respective standing wave. The net effect of the one-bit gate A_i on the i th qubit ($1 \leq i \leq N$) is given by

$$A_i |0\rangle_i = (|0\rangle_i + |1\rangle_i) / \sqrt{2}, \quad (14)$$

$$A_i |1\rangle_i = (|0\rangle_i - |1\rangle_i) / \sqrt{2}. \quad (15)$$

Also, two-bit quantum gates $B_{j,k}$ are needed, which couple the j th and k th qubits ($k > j$) in such a way that their product state is affected only if both qubits are in the upper level:

$$B_{j,k} |0\rangle_j |0\rangle_k = |0\rangle_j |0\rangle_k, \quad (16)$$

$$B_{j,k} |0\rangle_j |1\rangle_k = |0\rangle_j |1\rangle_k, \quad (17)$$

$$B_{j,k} |1\rangle_j |0\rangle_k = |1\rangle_j |0\rangle_k, \quad (18)$$

$$B_{j,k} |1\rangle_j |1\rangle_k = \exp\left(\frac{i\pi}{2^{k-j}}\right) |1\rangle_j |1\rangle_k. \quad (19)$$

It is much harder to realize this type of gate experimentally but it can be achieved in principle at least via a sequence of four standing wave pulses as shown by Cirac and Zoller in Ref. [1]. A common vibrational mode is used to couple the two ions, which are assumed to be in the vibrational ground state initially, with their equilibrium positions at the nodes of the respective standing waves. Each qubit receives a $\pi/2$

pulse (with zero laser phase), and then another $\pi/2$ pulse encoding the phase shift $\pi/2^{k-j}$ in the laser phase. These pulses have different laser polarizations for each qubit, and are detuned to the first red sideband so that the electronic and vibrational modes are coupled by a Jaynes-Cummings Hamiltonian. A vibrational mode independent from the mode for which the number distribution is measured has to be used in the implementation of these two-bit gates, in order to avoid disturbances to the vibrational statistics we want to measure. This additional vibrational mode could be in a direction perpendicular to the axis the ions are trapped on, and only its lowest two levels are used for the two-bit quantum gates.

Coppersmith's implementation of the quantum Fourier transform is equivalent to a classical discrete Fourier transform of the qubit amplitudes. The resultant network requires a number of quantum gates proportional to N^2 , however, more efficient schemes could be implemented using n -bit quantum gates, as pointed out by Cirac and Zoller [1]. The physical realization of this transform consists of a long sequence of standing wave pulses applied to qubits of the ion register, and can be represented by a series of quantum transformations (from right to left):

$$A_1 C_1 A_2 C_2 \cdots A_{N-1} C_{N-1} A_N, \quad (20)$$

where

$$C_r = B_{r,r+1} B_{r,r+2} \cdots B_{r,N}. \quad (21)$$

The order of the output qubits is reversed by this transformation, so that the i th qubit becomes the $[N - (i - 1)]$ th qubit in the electronic basis states $|k\rangle_e$. An approximate quantum Fourier transform has been proposed by Barenco *et al.* [10], and could be useful in reducing the number of two-bit quantum gates necessary to implement this transformation by some constant factor.

If the number of ions is large enough so that the vibrational distribution coefficients $|c_{k+2^N}|^2$ are negligible for $k \geq 0$, we can truncate the vibrational distribution from its 2^N th term to find an approximation to the final state of the register,

$$|\psi_{\text{out}}\rangle = \sum_{k=0}^{2^N-1} c_k |k\rangle_{\text{vib}} \otimes |k\rangle_e. \quad (22)$$

Hence we have obtained an approximate QND measurement of the initial vibrational number state distribution for a sufficiently large number of ions.

A readout of the ion register equivalent to measuring the observable Y can be made very efficiently using the quantum jumps technique [2,3] for every ion. Each of the two-level qubit transitions coupled to a standing wave is very strongly coupled to a third probe level by an intense resonant laser field. This three level scheme is the standard means of monitoring weak transitions in trapped ions. Since the upper levels of the qubits are metastable, we can determine if a particular ion is in the ground or the excited state by observing whether the resonance fluorescence from the probe level is on or off, respectively.

For large N , the probability of finding the ions in the state $|k\rangle_e$ is the same as the probability of the oscillator number state $|k\rangle_{\text{vib}}$ being occupied. Therefore the initial vibrational number state distribution can be determined from the relative frequencies of the electronic number states, taking care that the initial state of the system is reprepared accurately after each probe fluorescence measurement.

III. DISCUSSION AND CONCLUSION

The number of ions required to measure the initial vibrational number state distribution accurately is relatively small, since we truncate the distribution from its 2^N th term to obtain an approximate QND measurement in Eq. (22). For example, if all the terms $|c_n|^2$ of the vibrational distribution are

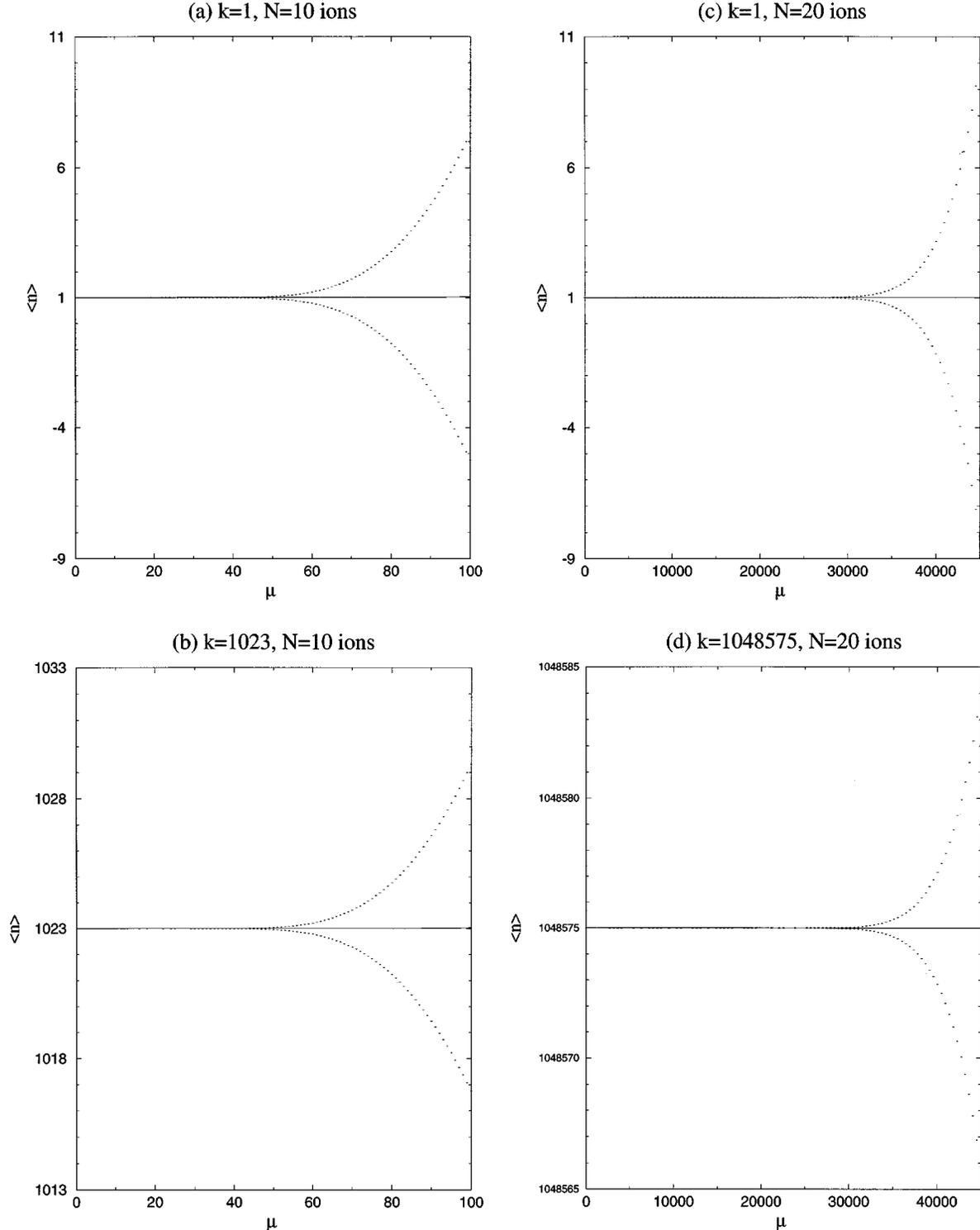


FIG. 1. Plots of the mean $\langle n \rangle$ (solid line) and uncertainties $n \pm \Delta n$ (dotted lines) of the phonon number in the conditional state $|\Psi(k)\rangle_{\text{vib}}$ against the mean phonon number μ of an initial thermal vibrational distribution for (a) $k=1$, $N=10$ ions, (b) $k=2^{10}-1$, $N=10$ ions, (c) $k=1$, $N=20$ ions, (d) $k=2^{20}-1$, $N=20$ ions.

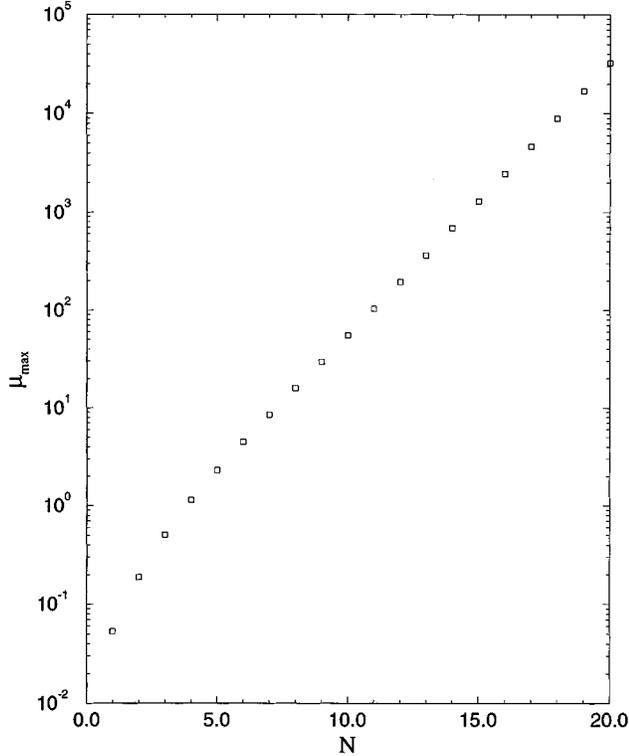


FIG. 2. Plot of the maximum value of the initial mean phonon number μ_{\max} for which the uncertainty in the mean phonon number $\Delta n \leq 0.1$, against the number of ions N ($k=1$).

negligible for $n > 10^{12}$, we need only 40 ions in the register to be able to determine all the important terms of the distribution. Currently the number of ions in a linear rf trap that can be trapped and manipulated in the Lamb-Dicke regime is limited [11], but we expect that it will be steadily increased by technological advances in ion trapping.

We now consider using an initial thermal vibrational state with our measurement scheme. The mean and variance of the phonon number were calculated numerically for the conditional vibrational state,

$$|\Psi(k)\rangle_{\text{vib}} = \sum_{n=0}^{\infty} \frac{c_{k+n2^N}}{\sqrt{\sum_{q=0}^{\infty} |c_{k+q2^N}|^2}} |k+n2^N\rangle_{\text{vib}}, \quad (23)$$

which is obtained if the ions are found in the state $|k\rangle_e$ using probe fluorescence measurements. For sufficiently large N , the conditional state reduces to a vibrational number state $|k\rangle_{\text{vib}}$, as expected for a QND measurement.

Figures 1(a)–(d) show the dependence of the mean phonon number $\langle n \rangle = \langle \Psi(k) | n | \Psi(k) \rangle_{\text{vib}}$ and its uncertainty Δn , on the initial mean phonon number μ of the thermal distribution. We have presented one case in which only one ion is in an excited state ($k=1$), and another where all the ions are in excited states ($k=2^N-1$), with $N=10, 20$ ions in the trap. Our results demonstrate that the uncertainty Δn increases exponentially with μ , but can be reduced below some acceptable level for a given value of μ by increasing the number of ions. The maximum value of the initial mean phonon number μ_{\max} for which the uncertainty Δn remains insignificant increases exponentially with N , as shown in

Fig. 2. Thus measurements of the vibrational number state distribution would be accurate for initial thermal states with mean phonon number $\mu \leq \mu_{\max}$. We note that given a fixed number of ions, the uncertainty Δn of the mean phonon number is the same for any value of k , as μ is increased.

Looking at our measurement scheme from a different point of view, we note that it can also be used to create many types of entangled ion states. The model we have outlined in this paper enables the preparation of an ion register in any superposition of electronic number states, if the corresponding initial vibrational state can be prepared. Recently, Meekhof *et al.* [12] have shown that a wide range of non-classical vibrational states can be prepared for a single trapped ion, and their methods could be extended to an N -ion system to provide initial vibrational states for our scheme. In particular, an initial vibrational number state can be used to load a given number into the ion register with unit efficiency.

In summary, we have shown in this paper that the initial vibrational number state distribution of an N -ion register can be determined using a set of far-detuned standing wave pulses and a series of quantum computations. Our scheme illustrates a general technique by which quantum computational transformations may greatly extend the class of operators accessible to measurement in quantum systems. Alternatively, our proposed measurement scheme can be used to create nonclassical entangled ion states.

APPENDIX

An interaction picture Hamiltonian H_I can be obtained from the Hamiltonian in Eq. (3) by assuming that $\eta_j \Omega_j / (2\sqrt{N}) \ll \Delta_j, \nu$ and applying a transformation $\exp[i(\sum_{j=1}^N \Delta_j \sigma_z^{(j)} + \nu a^\dagger a)t]$:

$$H_I = \frac{\hbar}{2\sqrt{N}} \sum_{j=1}^N \Omega_j (\sigma_+^{(j)} e^{i\Delta_j t} + \sigma_-^{(j)} e^{-i\Delta_j t}) \times \sin[\eta_j (a e^{-i\nu t} + a^\dagger e^{i\nu t})]. \quad (A1)$$

We assume that the detuning frequencies are all equal, i.e., $\Delta_j = \Delta$, and consider a Dyson expansion of the time evolution operator $U(t)$,

$$U(t) = 1 - \frac{i}{\hbar} \int_0^t H_I(t') dt' - \frac{1}{\hbar^2} \int_0^t \int_0^{t'} H_I(t') H_I(t'') dt'' dt' + \dots, \quad (A2)$$

generated by the Hamiltonian H_I .

The interaction Hamiltonian $H_I(t')$ consists of a sum of terms that are proportional to $\exp[i(\pm\Delta + p\nu)t']$ where $p = \pm 1, \pm 3, \pm 5, \dots$. All these terms average to zero for interaction times $t \gg \nu^{-1}$ if the detuning is well away from all vibrational resonance frequencies, i.e., $\min|\pm\Delta + p\nu| \approx \nu$, hence the first-order contribution to $U(t)$ in Eq. (A2) is zero.

Similarly, after calculating $\int_0^t H_I(t'') dt''$ the integrand of the second-order contribution to $U(t)$ consists of a sum of terms that are proportional to $\exp[i(\pm\Delta + p\nu)t']$, $\exp[iq\nu t']$, and $\exp[i(\pm 2\Delta + q\nu)t']$, where $p = \pm 1, \pm 3, \pm 5, \dots$ and $q = 0, \pm 2, \pm 4, \dots$. Given that the detuning is well away

from all vibrational resonance frequencies, as above, most of these terms also average to zero for interaction times $t \gg \nu^{-1}$. The nonzero terms are those proportional to $\exp(iqvt')$ and $\exp[i(\pm 2\Delta + qv)t']$ in the case that $q=0$, or $q=2\Delta/\nu$, respectively.

Assuming that all the Lamb-Dicke parameters are of the same order of magnitude, and the ions are in the Lamb-Dicke limit ($\eta_j \sim \eta_k \ll 1$), the contribution of the remaining terms can be ranked according to their order in η_j . The terms proportional to the lowest order of the Lamb-Dicke parameters dominate, and thus the second-order contribution to $U(t)$ in Eq. (A2) is given by

$$-\frac{i}{\hbar} \int_0^t dt' \frac{\hbar}{4N} \sum_{j=1}^N \sum_{k=1}^N \eta_j \eta_k \Omega_j \Omega_k \times \left(\frac{\sigma_+^{(j)} \sigma_-^{(k)} a a^\dagger - \sigma_-^{(j)} \sigma_+^{(k)} a^\dagger a}{\Delta - \nu} + \frac{\sigma_+^{(j)} \sigma_-^{(k)} a^\dagger a - \sigma_-^{(j)} \sigma_+^{(k)} a a^\dagger}{\Delta + \nu} \right). \quad (\text{A3})$$

The terms neglected in the Lamb-Dicke limit include all those terms resulting from the case where $q=2\Delta/\nu$ since we

assume that the detuning frequency is much greater than the trap frequency; i.e., $|\Delta| \gg \nu$. Using this approximation for the detuning, the time evolution operator can be rewritten as

$$U(t) \approx 1 - \frac{i}{\hbar} \int_0^t dt' \frac{\hbar}{4N\Delta} (2a^\dagger a + 1) \sum_{j=1}^N \sum_{k=1}^N \eta_j \eta_k \Omega_j \Omega_k S_{jk}, \quad (\text{A4})$$

where $S_{jk} = \sigma_+^{(j)} \sigma_-^{(k)} - \sigma_-^{(j)} \sigma_+^{(k)}$. The electronic operator S_{jk} is antisymmetric with respect to its indices,

$$S_{jk} = \begin{cases} -S_{kj} & (j \neq k) \\ 2\sigma_z^{(j)} & (j = k), \end{cases} \quad (\text{A5})$$

hence the off-diagonal terms ($j \neq k$) in Eq. (A4) are zero, and the Hamiltonian generating the time evolution operator $U(t)$ is given by

$$H_u = \frac{\hbar}{N\Delta} (a^\dagger a + \frac{1}{2}) \sum_{j=1}^N \eta_j^2 \Omega_j^2 \sigma_z^{(j)}. \quad (\text{A7})$$

After resetting the zeros of the vibrational and atomic energies, we obtain the Hamiltonian given in Eq. (4).

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