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Nonlinear coupling of continuous variables at the single quantum level

C. F. Roos,[1,2,*] T. Monz,[1] K. Kim,[1] M. Riebe,[1] H. Häffner,[1,2] D. F. V. James,[3] and R. Blatt[1,2]
[1]*Institut für Experimentalphysik, Universität Innsbruck, Technikerstraße 25, A-6020 Innsbruck, Austria*
[2]*Institut für Quantenoptik und Quanteninformation der Österreichischen Akademie der Wissenschaften, Technikerstraße 21a, A-6020 Innsbruck, Austria*
[3]*Department of Physics and CQIQC, University of Toronto, 60 St. George Street, Toronto, Canada M5S 1A7*
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We experimentally investigate nonlinear couplings between *individual quanta* of the vibrational modes of strings of cold ions stored in linear ion traps. The nonlinearity is caused by the ions' Coulomb interaction and gives rise to a Kerr-type interaction Hamiltonian $H=\hbar\chi\hat{n}_r\hat{n}_s$, where $\hat{n}_r$, $\hat{n}_s$ are phonon number operators of two interacting vibrational modes. We precisely measure the resulting oscillation frequency shift and observe a collapse and revival of the contrast in a Ramsey experiment over a range of trap frequencies. With 2.97 MHz stretch mode and 3.61 MHz rocking mode frequencies, we observed a coupling of 20.5 Hz/phonon. Implications for ion trap experiments aiming at high-fidelity quantum gate operations are discussed.

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Nonlinear interactions at the single quantum level have long been an elusive grail in the development of quantum technologies. In quantum optics, the implementation of a system with a Kerr nonlinearity (e.g., the Hamiltonian $H=\hbar\chi\hat{n}_r\hat{n}_s$) coupling two field modes will enable quantum-nondemolition measurements of single photons [1], the realization of quantum gate operations in photonic quantum computation [2–5], and photonic Bell state detection [6]. Unfortunately, the strong nonlinearities required to observe these effects at the single quantum level are difficult to realize [7,8]. However, in trapped-ion quantum computing the joint vibrational modes of the ion string have an intrinsic nonlinearity which is considerably stronger than that for photons. In this Rapid Communication we investigate this phonon-phonon coupling theoretically and demonstrate it experimentally for single quanta, and discuss some of its implications for quantum information processing technology.

The normal modes of ions' oscillations, which arise due to the strong coupling of their vibrations afforded by the Coulomb interaction, are of vital importance for the control and manipulation of the *internal* degrees of freedom of the ions. Using precisely tuned laser pulses, atomic excitations can be coherently transformed into quanta ("phonons") of the oscillatory modes, by which means multi-ion quantum gates are realized and entanglement created [9]. The normal mode description naturally appears when the ion trap potential is modeled as a harmonic (pseudo-)potential and the mutual Coulomb interaction between the ions is linearized around the ions' equilibrium positions [10]. In this way, the collective ion motion is described by a set of independent harmonic oscillators with characteristic normal mode frequencies. Small deviations from this picture arise because of the nonlinearity of the Coulomb interaction giving rise to a cross coupling between the normal modes. While nonlinear Coulomb couplings between ions found lots of attention in ion trap experiments investigating order-chaos transitions [11,12], its effect in the normal mode regime has mostly gone unnoticed. Here, we are not interested in resonant mode-mode coupling [13] but rather in dispersive cross-Kerr effects leading to shifts of the normal mode frequencies [14]. In this paper, we show that the Coulomb nonlinearity gives rise to a Kerr-type Hamiltonian and we quantitatively measure the strength of the induced frequency shift. Besides its implications for the fidelity of entangling quantum gates, this phonon-phonon frequency shift offers the potential for translating the tantalizing possibilities already considered for photons into experimental reality using phonons.

For two cold ions of mass m and charge e held in an anisotropic harmonic potential characterized by trap frequencies $\omega_z, \omega_x=\omega_y\equiv\omega_\perp$, with $\omega_z<\omega_\perp$, the equilibrium positions of the ions are given by $\mathbf{r_i}=(0,0,\pm z_0)$, where $z_0=[e^2/(16\pi\epsilon_0 m\omega_z^2)]^{1/3}$. Separating the center of mass (COM) and the relative ion motion by introducing the coordinates $\mathbf{R}\equiv(X,Y,Z)=(\mathbf{r}_2+\mathbf{r}_1)/2$ and $\mathbf{r}\equiv(x,y,z)=(\mathbf{r}_2-\mathbf{r}_1)/2$ yields the potential energy

$$U=m\omega_z^2\left(Z^2+z^2+\frac{\omega_\perp^2}{\omega_z^2}(X^2+x^2+Y^2+y^2)+\frac{2z_0^3}{r}\right).$$

Assuming the center of mass is stationary ($\mathbf{R}=0$), expanding the Coulomb potential to fourth order around $(0,0,z_0)$ by setting $z=z_0+u$ and keeping only those terms giving rise to a cross coupling between normal modes, we find

$$U\approx m\omega_s^2u^2+m\omega_r^2(x^2+y^2)+V^{(3)}+V^{(4)},$$

where $\omega_s=\sqrt{3}\omega_z$, $\omega_r=\sqrt{\omega_\perp^2-\omega_z^2}$ are the stretch and the rocking mode frequencies [15] and where $V^{(3)},V^{(4)}$ defined by

$$V^{(3)}=\frac{m\omega_s^2}{z_0}u(x^2+y^2),\quad V^{(4)}=-\frac{2m\omega_s^2}{z_0^2}u^2(x^2+y^2),$$

describe the cubic and quartic nonlinearities. Using these perturbation Hamiltonians and standard second-order perturbation theory we find the energy shifts $\epsilon(n_s,n_r^x,n_r^y)$ of the quantum states described by the quantum numbers (n_s,n_r^x,n_r^y), where n_s and $n_r^{x,y}$ specify the number of stretch mode and rocking mode phonons. The shift of the stretch

*christian.roos@uibk.ac.at

mode frequency by the rocking mode quantum numbers is given by

$$\delta\omega_s = \frac{\epsilon(n_s+1,n_r^x,n_r^y) - \epsilon(n_s,n_r^x,n_r^y)}{\hbar} = \chi(n_r^x + n_r^y + 1)$$

with the cross-Kerr coupling constant

$$\chi = -\,\omega_s\left(1 + \frac{\omega_s^2/2}{4\omega_r^2 - \omega_s^2}\right)\left(\frac{\omega_z}{\omega_r}\right)\left(\frac{2\hbar\omega_z}{\alpha^2 mc^2}\right)^{1/3}, \qquad (1)$$

where α denotes the fine-structure constant and c the speed of light. This expansion is valid as long as $\omega_r/\omega_s \gg (x_0/z_0)$ and $|2\omega_r - \omega_s|/\omega_s \gg (u_0/z_0)$, where x_0 (u_0) are the spatial half width of the rocking (stretch) mode ground states, respectively; in our experiments the spatial extent of the ion wave functions was always much less than the ions' separation, and the transverse trapping frequency was always considerably larger than the longitudinal frequency, thus these criteria are always met in practice, and Eq. (1) should be accurate.

In our experiments, two $^{40}Ca^+$ ions are confined in a linear Paul trap with radial trap frequencies of about $\omega_\perp/2\pi = 4$ MHz. By varying the trap's tip voltages from 500 to 2000 V, the axial center-of-mass frequency ω_z can be tuned from (2π)860 kHz to (2π)1720 kHz. The ions are Doppler cooled on the $S_{1/2} \leftrightarrow P_{1/2}$ transition. On the narrow $S_{1/2} \leftrightarrow D_{5/2}$ quadrupole transition, the vibrational sidebands are resolved [16]. This makes it possible to excite the ion either on the carrier transition without changing its vibrational state, or on the red (blue) sideband where the vibrational quantum number n is decreased (increased) to $n \pm 1$ upon excitation to the metastable $D_{5/2}$ state. Sideband cooling, which is akin to optical pumping of the population of the vibrational modes, using the $S_{1/2} \leftrightarrow D_{5/2}$ transition [16] prepares the stretch mode in the motional ground state $|0\rangle_s$. Simultaneous cooling of stretch and rocking modes is accomplished by alternating the frequency of the cooling laser exciting the quadrupole transition between the different red motional sidebands [15]. Motional quantum states are coherently coupled by a laser pulse sequence exciting a single ion on the $|S\rangle \equiv S_{1/2}(m=-1/2) \leftrightarrow |D\rangle \equiv D_{5/2}(m=-1/2)$ transition with a focused laser beam on the carrier and the blue sideband. Internal state superpositions $(|S\rangle + e^{i\phi}|D\rangle)|0\rangle$ can be mapped to motional superpositions $|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$ by a π pulse on the blue motional sideband and vice versa [17]. We discriminate between the quantum states $S_{1/2}$ and $D_{5/2}$ by scattering light on the $S_{1/2} \leftrightarrow P_{1/2}$ dipole transition and detecting resonance fluorescence of the individual ions with a charge-coupled device (CCD) camera. A more detailed account of the experimental setup is given in Refs. [17,18].

For a measurement of the stretch mode coherence, a Ramsey experiment between motional states is performed by a laser interacting with only one of the ions, the second ion being just a spectator that modifies the normal mode structure. Starting from the state $|S\rangle|0\rangle$, the superposition state $|D\rangle(|0\rangle + |1\rangle)$ is created by a $\pi/2$ pulse on the carrier transition followed by a π pulse on the blue sideband. During a waiting time of duration τ, the state evolves into $|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$, where ϕ is a random variable. Finally, the motional superposition is mapped back to superposition of

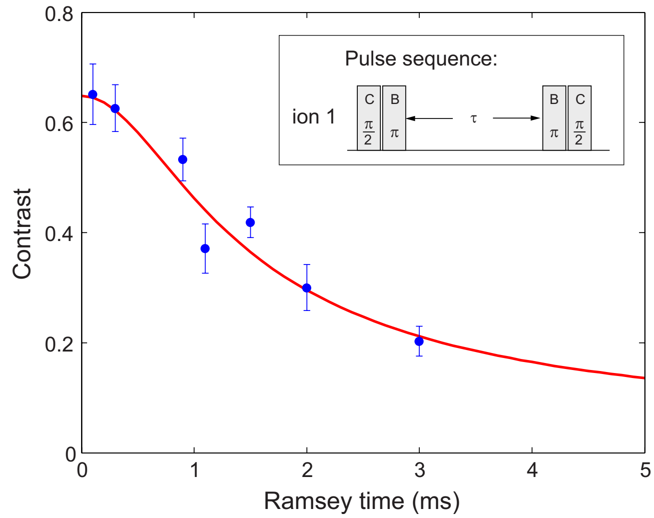

FIG. 1. (Color online) Contrast $C(\tau)$ of a Ramsey experiment on the stretch mode as a function of the Ramsey waiting time τ. The axial trap frequency was $\omega_z = (2\pi)$1486 kHz; the radial modes were cooled close to the Doppler limit. The initial contrast is limited by imperfect sideband cooling, spurious excitation of the second ion by the focused laser beam, and magnetic field noise. The subsequent loss of contrast is caused by a dephasing of the stretch mode oscillation. The inset shows the pulse sequence of the Ramsey experiment with C denoting carrier pulses and B blue sideband pulses.

internal states $(|S\rangle + e^{i\phi}|D\rangle)|0\rangle$ and probed by a $\pi/2$ pulse on the carrier transition followed by a quantum state measurement. We measure the coherence by varying the phase of the last $\pi/2$ pulse from 0 to 2π and measuring the contrast $C(\tau)$ of the resulting Ramsey pattern as a function of τ. The measurement shown in Fig. 1 exhibits a strong reduction in contrast after only 2 ms. This fast decay could be attributed neither to motional heating (because the stretch mode heating rate was less than 0.02 phonons/ms), nor to an instability of the trapping potential (since a coherence time of about 50 ms was measured on the center-of-mass mode). To probe the dynamics of the dephasing mechanism, a spin echo experiment was performed on a motional superposition $|0\rangle + |1\rangle$. For this, the populations of the $|0\rangle$ and $|1\rangle$ states were exchanged in the middle of the experiment by a carrier π-pulse sandwiched between two blue sideband π pulses. As can be seen in Fig. 2, now it takes the spin echo contrast $C(\tau)$ about 100 ms to decay to 50% of its initial value—a clear indication that the stretch mode frequency is fairly stable over the duration of a single experiment but randomly changing from experiment to experiment. This behavior is expected for a dephasing caused by a thermally distributed rocking mode phonon number that takes on random values at the start of each experiment. To confirm this dephasing mechanism, we remeasured the Ramsey contrast $C(\tau)$ with both rocking modes cooled close to the ground state and observed a 20-fold increase of the coherence time as compared to the experiment shown in Fig. 1.

Having established the cross coupling between the stretch mode and the rocking mode as the dephasing mechanism, we are interested in quantifying the cross-coupling strength by measuring the stretch mode frequency shift caused by a

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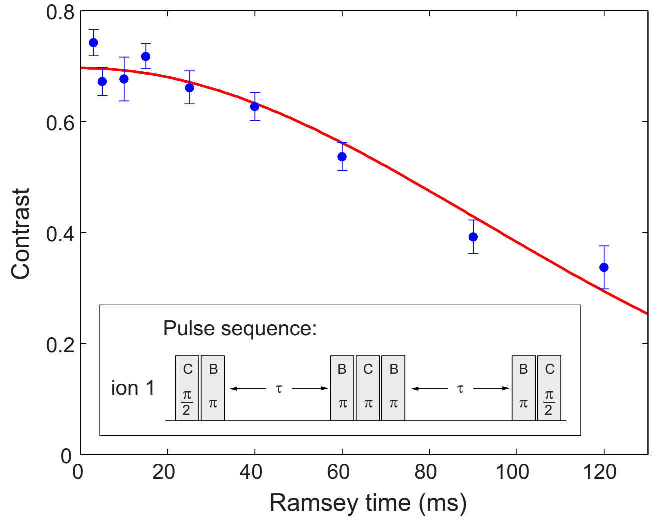

FIG. 2. (Color online) Contrast $C(\tau)$ of a spin echo experiment on the stretch mode as a function of the Ramsey waiting time τ. Insertion of the spin echo increased the coherence time by nearly two orders of magnitude as compared with a simple Ramsey experiment.

single rocking mode phonon. In principle, such a measurement could be performed by preparing the rocking modes in an $n_r=0$ or $n_r=1$ Fock state and subsequently measuring the stretch mode oscillation frequency in a Ramsey experiment. This, however, would require ultrastable high-voltage sources for keeping the axial trap frequency ω_z stable to within 10^{-6} ω_z or less. To make the experiment more robust against technical noise, we carried out a spin echo experiment instead, where the other ion was excited on the blue sideband of one of the rocking modes directly before the start of the second spin echo waiting time τ (see inset of Fig. 3). If this pulse excites the second ion into the $D_{5/2}$ state, there is exactly one additional phonon in the rocking mode during the second half of the spin echo whose frequency shift is not compensated by the echo sequence. We adjust the duration of this blue sideband pulse so that the excitation is successful in 50% of the experiments and sort the spin echo data into two classes according to the quantum state of the second ion at the end of the experiment. By measuring the phase shift $\Delta\phi(\tau)$ between the two data sets, we are able to infer the shift caused by a single rocking mode phonon. This procedure does not even require cooling the rocking modes to the ground state. Figure 3 shows the resulting phase shift for a waiting time $\tau=30$ ms. For a calculation of the single phonon frequency shift $d\nu_s/dn_r$, we plot the phase shift as a function of the spin echo time τ, fit a straight line to the data, and determine its slope. For the data measured at $\omega_z/2\pi = 1716$ kHz shown in Fig. 4 (inset), the frequency shift is $d\nu_s/dn_r=20.5$ Hz/phonon. This measurement procedure was carried out for different axial center-of-mass frequencies ω_z while keeping the transverse oscillation frequency $\omega_\perp$ fixed. In Fig. 4 the frequency shift is plotted as a function of ω_z. The experimentally measured frequency shift is somewhat smaller than the shift predicted by perturbation theory. The disagreement between the experimental data and theoretical model is currently not understood. It cannot be attributed to

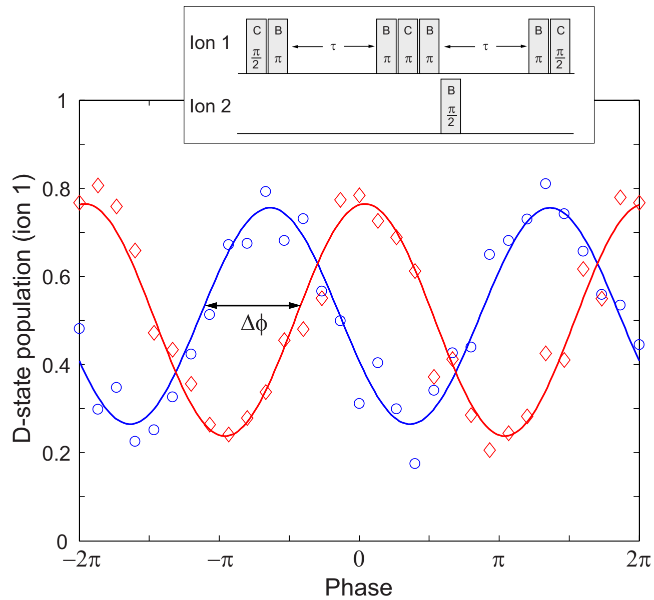

FIG. 3. (Color online) D-state population as a function of the spin echo phase. The shift $\Delta\phi$ of the spin echo phase pattern by a rocking mode phonon is observed by measuring the D-state population of ion 1 as function of the phase of the last spin echo pulse. The symbol (◇) labels experiments with an extra phonon in the second spin echo time, (○) experiments with no extra phonon.

the dynamical nature of the trap potential. Using a refined pseudopotential [19] leads only to marginal corrections.

For a thermally occupied rocking mode, the change induced in ω_s is always an integer multiple of the single phonon shift. Therefore, after the initial collapse of the Ramsey contrast (Fig. 1) a revival [20] is to be expected for a time

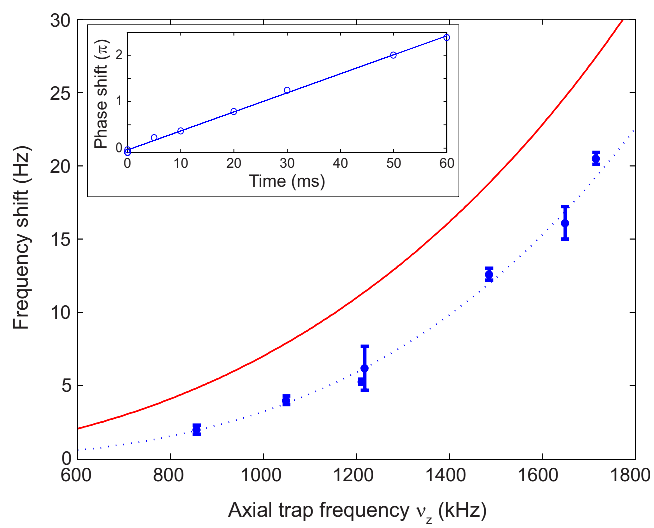

FIG. 4. (Color online) (Inset) Shift of the stretch-mode echo phase pattern as a function of the waiting time due to a single rocking mode phonon. The frequency shift is inferred from the slope of the function. (Main graph) Shift of the stretch mode frequency by a single rocking mode phonon as a function of the axial trap frequency ω_z. The solid line is the frequency shift predicted by Eq. (1); the dotted line is a fit to the data using a power law $d\nu_s/dn_r \propto \omega_z^{\beta}$ with $\beta=3.25$.

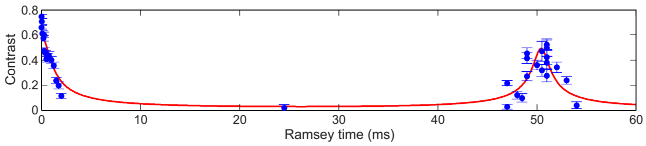

FIG. 5. (Color online) Collapse and revival of the contrast in a Ramsey experiment probing the motional coherence of the two lowest stretch mode quantum states. The revival occurs at the time predicted by the spin echo experiment for a frequency $\omega_z/2\pi = 1716$ kHz.

$\tau^* = 2\pi/\chi = 2\pi(d\omega_s/dn_r)^{-1}$. Operating the trap at $\omega_z/2\pi = 1716$ kHz, we indeed observed a revival at the predicted time τ^*. In this experiment, one of the rocking modes was sideband cooled to the ground state while the other one was prepared in a thermal state with $\bar{n}_r \approx 9$ (1σ confidence interval $\{5,17\}$) by Doppler cooling. Figure 5 shows a fit to the data using the function

$$\tilde{C}(\tau) = e^{-\gamma\tau}|\langle e^{i\chi\hat{n}_r\tau}\rangle| = e^{-\gamma\tau}|\bar{n}_r + 1 - \bar{n}_r e^{i\chi\tau}|^{-1},$$

assuming a thermally occupied rocking mode and an overall loss of contrast $\propto e^{-\gamma\tau}$ accounting for technical noise and motional heating. A fit to the data yields a revival time $\tau^* = 50.5(5)$ ms, an average vibrational quantum number $\bar{n}_r = 9(2)$ that is consistent with an independent measurement of $\bar{n}_r$ and a decay rate $\gamma = 0.004(3)$ s^{-1}. The existence of revivals is a further proof of the quantized nature of the rocking motion. The small loss of contrast shows that the probability of a change in the rocking mode phonon number within the interval $[0,\tau^*]$ is quite low.

The observed frequency shifts need to be taken into account in quantum gate realizations operating on the stretch mode. For the parameters [21] used in Ref. [9], Eq. (1) predicts shifts as big as 100 Hz/phonon, giving rise to a loss of fidelity of about 0.1% for $\bar{n} = 1$, which will be of significance as the fault-tolerance threshold for quantum computers is approached in the years ahead. Moreover, the gate realizations demonstrated in [18] are believed to have been affected by cross-mode coupling at the percent level.

In summary, we have investigated a nonlinear quantum effect giving rise to a cross coupling of harmonic oscillators that can be described by a Kerr-like Hamiltonian $H \propto \hat{n}_s\hat{n}_r$. In a two-ion crystal, the nonlinearities lead to a dephasing of the relative ion motion that manifests itself as a collapse of the Ramsey contrast that revives once all oscillations go in phase again. While the nonlinearity is fairly small for the trap parameters investigated here, it could be made bigger by tuning the normal mode frequencies closer to the resonance $\omega_r = 2\omega_s$ so that it might be used for creating entangled motional states of these oscillators. On the other hand, for quantum computing experiments aiming at fault-tolerant quantum gates, the observed nonlinearity points to the necessity of cooling all spectator modes to the ground state or working with transversally very stiff ion traps.

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