Measurement-Induced Vibrational Dynamics of a Trapped Ion

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Excitation spectra of an individual trapped $138Ba⁺$ ion show sidebands of radial and axial ion vibration in the trap. The second-order sidebands involving radial *and* axial vibrations would allow swapping these vibrational states; they represent an inverse Raman effect with the role of electronic and vibrational excitation interchanged. Deexcitation sidebands differ from excitation sidebands as the vibrational distribution in the $D_{5/2}$ level is modified by the ion being reduced to this level in null observations of resonance light. Delayed deexcitation goes along with stochastic cooling that makes the ion collapse into the Fock state $|n = 1\rangle$. Sideband modulation is identified as stroboscopic detection of the light-induced nutation on a vibrational transition with $\Delta n = \pm 1$. [S0031-9007(98)05673-7]

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Individual atomic particles—ions—have been prepared in an rf trap and observed by their laser-excited resonance light for quite a while [1,2]. Trajectories of the evolution of an electronic degree of freedom have been recorded in this way and have revealed Bohr's "quantum jumps" [3,4]. The ion's center-of-mass motion has been almost eliminated [5,6]. Complete command of the ion's center-of-mass motion is prerequisite when one intends to use one of the narrow, unshifted ion resonances as a frequency mark for the long-term control of an rf or laser oscillator thus turning the combination into a highaccuracy frequency standard [7]. Recently, individually light-controlled ions have been suggested to serve as logic gates for quantum computation [8], and the steps of manipulation required for implementation of a "controlled not gate" have been demonstrated, making use of both the ground-state hyperfine transition, and the vibrational excitation of a ${}^{9}Be^+$ ion [9].

The center-of-mass motion of an ion in an rf trap consists of free harmonic secular vibrations superimposed on the oscillatory "micromotion" driven by the rf field. Since this field displays a node at trap center, the micromotion is minimized by suitably positioning the ion [10,11]. The vibration is reduced by laser cooling on a resonance line ("Doppler" cooling) [12,13]; even somewhat more by Raman cooling [14,15]. Reduction to the vacuum state has required more complicated schemes [5], and preparation and manipulation of particular states of the vibrational motion have been demonstrated [16]. Application of these techniques to implementing more complex logic gates requires, however, extra memory qubits not available so far.

We report on the simultaneous controlled laser excitation of a dipole-forbidden optical transition and a secular vibration of a single $138Ba⁺$ ion. We demonstrate *two* orthogonal vibrational motions [17] that must become anticorrelated upon excitation of a second-order vibrational sideband close to the carrier line, such that the setting of the states of vibration might be interchanged with no *a priori* knowledge of the information coded in the set-

ting. Moreover, we show, for the first time, that the very procedure of driving and probing the ion by light pulses (i) *modifies* the ion's *vibrational distribution* and (ii) will eventually amount to *dynamic preparation* of the ion either in a vibrational Fock state or in a stationary or timevarying superposition of such states. The former modification is detected in the strength of the sidebands and evidences stochastically cooling the ion, a filtering process in momentum space that has been recently predicted [18]. The process is shown to make the ion *asymptotically* approach a Fock state. It goes on, since the driving light's effect on the ion vibration remains coherent as long as the ion is not actually deexcited and the probe light gets resonantly scattered.

In the experiment, a $Ba⁺$ ion is loaded into a 1-mmsized radio frequency trap, and irradiated by laser light at 493 and 650 nm, for excitation of resonance scattering on the $S_{1/2}$ - $P_{1/2}$ line and prevention of optical pumping into the $D_{3/2}$ level, respectively. The ion is laser cooled by the green light down tuned from resonance. The fluorescence signal (3 \times 10⁴ s⁻¹ maximum rate) is recorded by photon counting. So far, the setup has been described elsewhere [11]. Light of a tunable color-center laser $(NaCl:OH^-)$ at 1.762- μ m output of 15 kHz bandwidth excites the ion on its $E2$ line $S_{1/2}$ - $D_{5/2}$ [6]. In fact, irradiation by this light alternates with that by the green probe light, and absence (presence) of resonance scattering is the signature of the ion having been excited to the $D_{5/2}$ (left in the $S_{1/2}$) level [19] (Fig. 1). The number of pairs of measurements with resonance scattering "on-off," normalized by the total number of measurements, represents the probability of excitation on the *E*2 line. A set of such measurements recorded upon stepwise scanning the frequency ω_l of the ir laser yields a spectrum of absorption [Fig. 2(a)]. Such spectra show sidebands from Doppler phase modulation by the ion's secular motion in the trap. The mean vibrational quantum number $\langle n \rangle$ may be derived provided that the distribution of the ion over the vibrational states is known. With thermal distribution, the strengths of upper and lower unsaturated first-order sidebands vary as $(\langle n \rangle + 1)\eta^2 \Omega^2$

FIG. 1. Laser excitation of the $Ba⁺$ ion on the $E2$ (quadrupole) line to its $D_{5/2}$ level, and on the resonance line $S_{1/2}$ -*P*_{1/2} line (top) alternate and form 4-ms-long cycles of measurement (bottom) whose result is either photocount rate "on" (ion is mostly in $S_{1/2}$) or "off" (ion in $D_{5/2}$).

and $\langle n \rangle \eta^2 \Omega^2$, respectively, where η is the Lamb-Dicke parameter, and Ω is the *E*2 Rabi frequency [20]. The ratio S_l/S_u of these strengths does not depend on the light intensity at the ion location, whereas it *does* for other distributions [21]. With a Fock state, e.g., this ratio approaches unity upon increasing Ω . However, after each probing of resonance scattering of the green light, the ion is supposed thermalized with $\langle n \rangle = S_l/(S_u - S_l)$ as, e.g., in Fig. 2(a), where $\langle n \rangle = 3.2$.

In fact, under better resolution the ion displays two vibrational modes (ω_r, ω_z) , and its excitation spectrum shows two sets of sidebands, since the potential of the trap is spheroidal, and ω_r , ω_z are nondegenerate. Figure 2(b) shows such a spectrum, where pairs of first-order sidebands are resolved. The carrier line ("zero-phonon" line in Mössbauer terminology) and the radial sidebands are strongly saturated, the axial ones weakly. Close to the carrier line, unsaturated second-order sidebands show up that result from the ion simultaneously exchanging a radial and an axial vibrational quantum, i.e.,

$$
\omega_l \equiv \omega_0 + \Delta_{ir} = \omega_0 \pm (\omega_z - \omega_r),
$$

where ω_0 is the *E*2 resonance frequency. The energy defect of these vibrational quanta is made up by the offset of the light from resonance. This situation is opposite to the one encountered in stimulated Raman excitation, where the energy difference of *electronic* pump and Stokes quanta is made up by the *vibrational* excitation. Consequently, this type of fundamental atomlight interaction qualifies as an *inverse Raman* process. With the combined excitation of an internal resonance and a vibrational degree of freedom both serving as qubits

FIG. 2. (a) Excitation spectrum of the *E*2 line $S_{1/2}$ - $D_{5/2}$
 $(m_S = +1/2 \rightarrow m_D = +3/2)$ whose (first-order) side- $(m_S = +1/2 \rightarrow m_D = +3/2)$ whose (first-order) side-
bands are compatible with $\langle n_r \rangle + \langle n_r \rangle = 3.2$. Laser bands are compatible with $\langle n_r \rangle + \langle n_z \rangle = 3.2$. bandwidth 50 kHz; sidebands unsaturated. (b) Excitation spectrum with *r* and *z* vibrational sidebands resolved, and *r* sideband and central line strongly saturated. The latter is accompanied by second-order sidebands that mark light-induced interchange of *r* and *z* vibrational quanta ("inverse Raman effect"). Bandwidth 15 kHz. (c) Deexcitation spectrum showing equal first-order sidebands twice as large as in (a). Bandwidth $= 50$ kHz; sidebands unsaturated.

for the operation of a quantum-logic gate, the second vibrational degree of freedom would provide the ion with an extra memory qubit. Excitation of those secondorder sidebands would allow one to shift the ion from a particular state of the *r* vibration to the corresponding one of the *z* vibration or vice versa [22].

After the ion has been excited, on a sideband, to the $D_{5/2}$ level, its vibrational distribution is nonthermally bunched around a value of $\langle n \rangle$ that is higher than before, in the $S_{1/2}$ ground state [17]. Triples of measurements with the result of probing—i.e., the resonance light found "on-off-on" represent events of prompt stimulated *deexcitation*. The spectra derived from these events show both upper and lower first-order sidebands varying as $(2\langle n \rangle + 1)\eta^2 \Omega^2$ [Fig. 2(c)]. Moreover, there are events with more than one "off" result sandwiched between initial and final "on" observations. These data correspond to delayed deexcitation. Sequences of such null detections have been shown to prepare the ion in a state of lower $\langle n \rangle$, i.e., to stochastically cool the ion [18]. In fact, $\langle n \rangle$ is supposed to decrease monotonously upon increasing number *q* of intermediate off results in consecutive measurements. This number represents the length of time the metastable ion undergoes intermittent interaction with the driving light but fails to get actually deexcited to its ground state. Figure 3(a) shows deexcitation spectra, plotted vs *q*, that represent the probability of deexcitation

immediately after *q* futile attempts. This probability

$$
W(q) = -dU(q)/dq
$$

decreases since uninterrupted sequences of *q* null detections turn rarer upon *q* growing. Here

$$
U(q) = \prod_{p=1}^{q} \rho_{11}(\tau, p)
$$

is the probability for finding *q* sequential off results, where $\rho_{11}(\tau, p) = \sum_n \rho_{11}^n(\tau, p)$ is the ion's occupation density of the $D_{5/2}$ level, and τ is the duration of the interaction with the driving light [17,18]. In Figs. 3(b) and 3(c), this probability has been both calculated for the initial thermal vibrational distribution determined by the absorption spectrum and derived from the sidebands of radial vibration of Fig. 3(a): Deexcitation events on the carrier line ($\omega_l = \omega_0$, left plot) do not change the vibrational state of the ion; such events on the (upper) sideband do $(\omega_l = \omega_0 + \omega_r$, right plot). Moreover, in sideband deexcitation the probability *U* much exceeds a corresponding result of computation that neglects renormalization of ρ_{11} after measurements that fail detecting resonance light [dashed line, Fig. $3(c)$]. The relative excess of probability growing with *q* and time reflects the matrix element for light interaction on the vibrational sideband diminishing upon decreasing $\langle n \rangle$, and thus it provides indirect evidence for stochastic cooling of the ion.

The sidebands of *axial* vibration in Fig. 3(a), being just weakly saturated, are more susceptible to any variation of the transition rate. When plotting the ir deexcitation probability $\left(-\frac{dU}{dq}\right)/U = P_{\text{decay}}$ vs *q*, we notice this probability decreasing, i.e., direct evidence of stochastic cooling, and also modulation with temporally increasing contrast [Fig. 4(a)]. This modulation of the deexcitation probability during sequences of measurements that lack actual deexcition characterizes the vibrational evolution of the ion as coherent dynamics. The corresponding probability of deexcitation is [16]

$$
P_{\text{deex}} = \frac{1}{2} - \frac{1}{2} \sum_{n=0}^{+\infty} Q_n \cos(2t \Omega_{n,n+1}),
$$

where $Q_n = \rho_{00}^n + \rho_{11}^n$ is the population of vibrational where $Q_n = \rho_{00} + \rho_{11}$ is the population of vibrational level *n*, and $\Omega_{n,n+1} = \eta \Omega \sqrt{n+1}$ are the Rabi nutation frequencies related to the upper-sideband vibrational transition $|n\rangle \rightleftharpoons |n + 1\rangle$, and Ω is the optical Rabi frequency determined by the *E*2 matrix element and the light amplitude at the ion location. If the ion's vibrational distribution peaks at $\langle n \rangle$, a good approximation is

$$
P(S_{1/2}, \langle n \rangle) = P_{\text{deex}}(q) \approx \sin^2 q \tau \Omega_{\langle n \rangle, \langle n \rangle + 1}.
$$

Since driving the *E*2 transition alternates with probing on the resonance line, the nutation dynamics between vibrational states $|n\rangle$ and $|n \pm 1\rangle$ goes on during the first half of each cycle of measurement, but stops for the second half. When no resonance light is detected within this probing half cycle, the ion is found to reside in the $D_{5/2}$ level, and its vibrational state including

FIG. 3. Number of emission events that form deexcitation spectra $W(\Delta_{ir})$ vs number *q* of "off" results (a). Probability $U(q)$ for *q* sequential null detections of resonance light: At the central line [vibration unaffected, (b)], at the upper sideband ω_r (c) where vibration and probability of deexcitation decrease such that $U(q)$ exceeds corresponding value calculated with modification of vibrational distribution neglected (dashed line).

its phase of nutation acquired by the previous action of the driving light remains unaffected by the probe light [Fig. 4(b)]. This nutational phase varies stepwise and periodically over such a sequence of cycles as long as neither stimulated nor spontaneous deexcitation terminates the sequence. Eventually, probing for the ion's ground state may turn successful, and resonance light will be detected. The probability of this event happening depends on the achieved nutational phase and, therefore, periodically on *q*: The light-driven nutation, alternating with intervals of free vibration during "null" probing, is *stroboscopically* recorded. The stroboscopic period is infinite if the duration τ of the driving-light irradiation is an integer multiple *m* of the nutation period $\tau_N =$ $2\pi/\Omega_{\langle n \rangle, \langle n \rangle+1}$; in general, it varies as $|\tau - m \cdot \tau_N|^{-1}$, and the stroboscopic frequency is

$$
\nu_s = (\Omega/2\pi)(\tau - m\tau_N)/\tau.
$$

The data in Fig. 4(a) are compatible with $2\pi\nu_s \approx \Omega/50$ and $m = 5$. Note that the contrast of modulation

FIG. 4. (a) Peak values on sidebands are modulated, vs *q* or time, by coherent vibrational ion dynamics during uninterrupted null detections. At $q = 0$ and 20, $\langle n \rangle = 109$ and 7.9, respectively, and it would be 1.3 at $q = 100$. (b) Laser-driven coherent vibrational ion dynamics adds, during each half-cycle τ of interaction, another increment $\Delta \theta = \tau \Omega_{\langle n \rangle, \langle n \rangle + 1}$ to the nutation phase $\theta = (q - 1)\Delta\theta$ which stays constant during half cycle of probing. θ is detected via $P_{\text{deex}}(q)$.

increases with *q* towards unity, and one Fourier component $\Omega_{n,n+1}$ emerges, since the ion *dynamically approaches* a Fock state of its vibration. With a long enough sequence of null measurements this state is supposed to be $|n = 1\rangle$. Initial excitation by light at the *carrier* frequency succeeded by frustrated attempts of deexcitation on the upper sideband would leave the ion in the "trapped" *vacuum* Fock state.

Individual trapped ions to be used as frequency marks for the control of standard oscillators or as logic gates for quantum computation require efficient cooling of the ions' center-of-mass motion as well as the preparation of pure motional states, in particular, of Fock states. The demonstrated procedure is well-suited for this purpose, since it is reliable and ready to apply. Moreover, stochastic cooling is about a thousand times faster than sideband cooling applied on the *E*2 line.

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