Laser sideband cooling with positive detuning

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An experimental and theoretical study of laser sideband cooling of a trapped ion in a radiofrequency trap is presented. The influence of the micromotion in the time-dependent potential of a Paul trap can lead to a counterintuitive situation, where sideband cooling is possible for positive values of the laser detuning, i.e., for a laser frequency that is higher than the resonance frequency of the ion at rest. The cooling rate and the capture range are calculated in a semiclassical model. Experimental results of laser sideband cooling at positive detuning are demonstrated with a single trapped In^+ ion stored in a miniature Paul trap.

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I. INTRODUCTION

The essence of almost all laser cooling methods [1] is that kinetic energy of the atoms is transferred to the photon field in anti-Stokes scattering processes [2]: On the average, the emitted photons are of higher frequency than the absorbed photons. This condition can be ensured in a two-level system by a negative or "red" detuning $\delta = \omega_L - \omega_0$ between the laser frequency ω_L and the resonance frequency ω_0 of the atom at rest. In Doppler cooling or optical molasses [3] the negative detuning is compensated by the Doppler shift for atoms that move opposite to the direction of the laser beam, so that the scattering of photons results in a velocity-dependent damping force. In sideband cooling of trapped atoms or ions [4] the negative detuning is usually chosen to match the vibrational frequency ω of the particle in the trap, $\omega_L = \omega_0 - \omega$, so that the excitation of the red sideband and anti-Stokes scattering prevails over nonresonant excitation of the blue sideband and Stokes scattering. In these simple pictures of Doppler or sideband cooling, a change of the sign of the detuning from negative to positive will reverse the cooling processes into heating.

Under special conditions, however, it is also possible to obtain laser cooling for positive or "blue" detuning and to satisfy the condition of anti-Stokes scattering even then. This has been demonstrated for neutral atoms in cases where the combination of a spatial modulation of the light shift produced by a blue detuned standing laser field and the associated modulation of the optical pumping rate leads to a realization of the Sisyphus myth: the moving atom has to climb uphill on light shift potentials most of the time and emits photons of a higher energy than it absorbs [5–9]. A combination of light shift and quantum interference with two blue detuned laser frequencies driving a three-level system has recently been used for efficient cooling of a trapped ion in a method called cooling with electromagnetically induced transparency [10].

In this paper we investigate laser sideband cooling for an ion trapped in the time-dependent potential of a radiofrequency trap and show experimentally that in this case laser cooling with positive detuning is possible in a two-level system. The standard description of laser cooling of trapped ions is limited to the case of a static harmonic trap [11–13]. However, the potential in a real Paul trap is not static but time dependent. The motion of an ion in a Paul trap [14,15] is composed of the so-called "secular motion" in the time-averaged harmonic pseudopotential and the micromotion at the frequency of the applied alternating electric field. We assume that the radiative atomic resonance linewidth γ is much smaller than the motional frequencies, so that modulation sidebands are well resolved in the optical spectrum (i.e., the so-called "strong-binding condition" is satisfied). The more complex motional spectrum can lead to significant modifications of the cooling dynamics in comparison with the predictions for laser sideband cooling in a static harmonic trap. In particular, the dependence of the ion's kinetic energy on the laser detuning becomes more intricate. This was first pointed out in a theoretical analysis by Cirac *et al.* [16], where a quantum mechanical calculation restricted to the Lamb-Dicke regime (LDR) was employed. These modifications were later confirmed by a semiclassical investigation valid also beyond the LDR [17]. Both studies agree in particular on the possibility of laser sideband cooling for laser frequencies above resonance, i.e., for positive detunings. This is in contrast to the results obtained within the standard theory of laser sideband cooling in a harmonic trap, where cooling occurs solely for laser frequencies below resonance [11–13]. For weakly bound ions with unresolved sidebands $(\gamma \gg \omega)$ a similar inversion of cooling and heating as a function of the laser detuning can occur under conditions of strong micromotion, as in a trapped Coulomb crystal [18,19].

In Sec. II, we review our semiclassical model [17] of sideband cooling in a radiofrequency trap and use it to determine the capture range of cooling for positive detuning. We present the first experimental result for the effect from an experiment with a trapped In^+ ion in Sec. III and compare the outcome with the semiclassical simulations.

II. SEMICLASSICAL MODEL

The motion of a particle of charge e and mass m in the quadrupole potential of a radiofrequency trap is described by

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a Mathieu differential equation. In the adiabatic approximation a truncated Floquet ansatz leads to stable solutions, which can be written in the form [15]

$$r(t) = r_0 \cos(\omega t)\left(1 + \frac{q}{2}\cos \Omega\, t\right). \quad (1)$$

Here $q = 2eU_0/m\Omega^2\rho^2$ is the dimensionless parameter that is determined by the charge and mass of the ion, the amplitude U_0 and the frequency Ω of the alternating voltage, and the radius ρ of the trap ring electrode. In most experiments, q is approximately 0.2. According to Eq. (1), the motional spectrum of the ion consists of three resonances: one at the secular frequency ω with an amplitude r_0 and two at the sum and difference frequencies $\Omega \pm \omega$, corresponding to the ordinary micromotion [20], with an amplitude that is a factor $q/4$ smaller than that of the ω term.

In the interaction with the laser light the frequency modulation due to the ion's motion will cause a comb of sidebands for each of the three frequencies. In addition to the carrier, the absorption spectrum will show sidebands at detunings $j\omega$, $j^{\pm}(\Omega \pm \omega)$, $(j, j^{\pm} = 0, \pm 1, \ldots)$. At a given laser detuning absorption can take place through sidebands of different order for different modulation frequencies. All processes that involve absorption on a low-frequency secular sideband $j < 0$ will lead to cooling; all those involving a high-frequency secular sideband $j > 0$ to heating. The change of the kinetic energy of the ion's secular motion that is produced through these various resonances is obtained from the product of the respective scattering rate and the detuning. In addition, each scattered photon increases the average energy of the ion by the photon recoil energy $E_R = \hbar R$, where R is the recoil frequency $\hbar k^2/2m$.

In the following, the ion is treated as an oscillator with amplitude r_0 at frequency ω and amplitudes $r_0 q/4$ at $\Omega \pm \omega$. The strength of a sideband corresponding to modulation order j of the secular motion, order j^+ of the modulation at the frequency $\Omega + \omega$, and order j^- of the modulation at the frequency $\Omega - \omega$ is given by the product of the squares of Bessel functions: $J_j^2(\beta) J_{j^+}^2(\beta q/4) J_{j^-}^2(\beta q/4)$, where $\beta = 2\pi r_0/\lambda$ is the modulation index of the secular motion. From the amplitude of the secular motion the mean secular kinetic energy $E = m r_0^2 \omega^2/2$ is calculated, which is expressed here in units of quanta of oscillation energy $\hbar\omega$.

If in addition a supplementary static force is present, due to, e.g., uncompensated static electric stray fields which shift the ion to a new equilibrium position r_1, the approximate solution (1) to the equation of motion is modified:

$$r(t) = (r_1 + r_0 \cos \omega t)\left(1 + \frac{q}{2}\cos\ \Omega\, t\right). \quad (2)$$

In this case, sidebands at multiples of the trap frequency Ω will also appear in the excitation spectrum. The modulation index of these sidebands is not linked to the amplitude of the secular motion as is the case for the modulation index of the ordinary micromotion: the amplitudes of the additional micromotion [20] sidebands thus stay constant and are not reduced by laser cooling. They can easily be incorporated into the model by an additional modulation of the laser at frequency Ω.

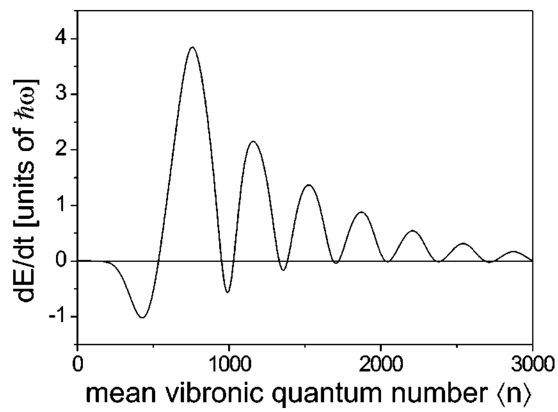

FIG. 1. Cooling rate as a function of the mean vibrational quantum number $\langle n \rangle$ for $\delta/2\pi = +7$ MHz (with $\omega/2\pi = 1$ MHz, $\Omega/2\pi = 10$ MHz) according to the semiclassical model. Cooling toward the vibrational ground state of the trap occurs as long as $\langle n \rangle < 500$.

For each value of the laser detuning, the absorption of photons from several sidebands—from both the secular and the micromotion of the ion—of different order may lead to competing cooling and heating processes. In the Lamb-Dicke regime a qualitative analysis is particularly simple because the sidebands of the lowest order will dominate. In this case efficient cooling is expected for detunings on the red side of each micromotion sideband and heating for detunings on the blue side of the micromotion sidebands. This result holds with or without additional micromotion at Ω due to a static displacement of the ion. The quantum mechanical calculation [16] showed that cooling at positive detuning may lead to similarly low kinetic energies as cooling at $\delta < 0$. The seemingly counterintuitive situation of an ion performing anti-Stokes scattering with laser photons that are already shifted to the blue side of the transition frequency is resolved by looking at the frequency modulated laser spectrum from the rest frame of the ion: From the comb of laser sidebands, the ion will preferentially select low-energy photons and will reemit these at anti-Stokes shifted frequencies.

In the limit of very large oscillation amplitudes—opposite to the Lamb-Dicke regime—many sidebands will have approximately equal strength. The predominance of cooling or heating will then be determined mainly by the sign of the detuning, as in the case of laser cooling in a static trap. Consequently, we expect the sideband cooling at blue detuning to be effective only over a limited range of oscillation amplitudes.

Figure 1 shows the result of the semiclassical model for the cooling rate as a function of vibrational energy. The parameters for the calculations were chosen to approximate those of our experiment: $q = 0.2$, $\Omega/\omega = 10$, $\gamma/\omega = 0.36$, $R/\omega = 0.032$, $\omega = 2\pi \times 1$ MHz. The laser intensity was assumed to be well above saturation ($I = 50\ I_{sat}$) and the detuning positive at $\delta/2\pi = +7$ MHz. Additional micromotion with a significant amplitude (modulation index ~ 2), analogous to the experimental situation (see Sec. III), has been incorporated in the simulation.

The oscillatory character of the Bessel functions used to calculate the strengths of the different sidebands leads to oscillations of the cooling rate as a function of the mean vibrational quantum number. The cooling curve shows several zero crossings and those with a negative slope represent stable states with capture ranges that are bounded by the adjacent zero crossings with positive slope. As already pointed out in [17], taking the micromotion into account, sideband cooling bears the effect of multistability. Cooling to the lowest temperatures is obtained only if the amplitude of the secular motion is initially sufficiently small to be within the capture range of the energetically lowest steady state. For the parameters of the simulation shown in Fig. 1, this holds for $\langle n \rangle < 500$. This is in contrast to detunings $\delta < 0$ where cooling can be efficient over wide ranges of the vibrational energy [17].

Decreasing the amplitude of additional micromotion in the simulation leads to a cooling rate with a smaller fraction of the curve above the zero line and so the lowest capture range for cooling at blue detuning becomes smaller. Therefore additional micromotion is beneficial for the experimental demonstration of cooling at positive detuning.

III. EXPERIMENT

Sideband cooling of In^+ is performed by excitation of the $5s^2\ {}^1S_0 - 5s5p\ {}^3P_1$ intercombination line with a natural linewidth $\gamma = 2\pi \times 360$ kHz [21]. This transition is narrow enough to meet the strong-binding criterion for common trap geometries, but is also sufficiently broad to enable fast and efficient laser cooling without the need for precooling the ion on a second, faster transition, even when starting from very high mean vibrational quantum numbers. A single laser source suffices for laser cooling, because the ground state has no hyperfine splitting and the transition represents a nearly closed two-level system with only a weak decay channel from the ${}^3P_1$ level toward the metastable $5s5p\ {}^3P_0$ state (decay rate $\approx 2\pi \times 35$ mHz [21]). With red detuned sideband cooling on the ${}^1S_0 - {}^3P_1$ line we have demonstrated reduction of the mean vibrational quantum number of a single ion from an initial $\langle n \rangle \sim 10^8$ to about $\langle n \rangle = 0.7$, corresponding to a temperature of $T \approx 60\ \mu$K [17].

The apparatus of our experiment has been described in detail elsewhere [17,22]. Here we give just a short overview of the basic setup. For trapping a single In^+ ion we use a miniaturized Paul-Straubel trap consisting of a copper beryllium ring electrode of 1 mm diameter. About 1000 V ac at 10 MHz and 30 V dc are applied to this electrode while zero potential is defined by two end plates placed at a distance of 4.5 mm from the trap. The corresponding secular frequencies are about 1.4 MHz in the axial and 0.9 MHz in the two radial directions.

As laser source a frequency-doubled stilbene-3 dye laser is used, which is frequency stabilized to an external reference cavity of finesse 3000, thus achieving a laser linewidth well below 10 kHz [17]. The laser frequency is tuned to the hyperfine component $F=9/2 \rightarrow F=11/2$ of the ${}^1S_0 - {}^3P_1$ transition. Via optical pumping with circularly polarized light in a vanishing magnetic field, a closed system of only two Zeeman sublevels is obtained. Laser cooling is effective in all three dimensions simultaneously because the laser wave vector has nonvanishing projections along all three major axes of the trap and the differences between the secular frequencies are smaller than or comparable to the natural linewidth of the transition. Therefore, irrespective of the precise value of the laser detuning, at least one sideband of each vibrational degree of freedom is always close to the laser frequency.

Figure 2 shows experimental excitation spectra of a single trapped In^+ ion, obtained by tuning the laser over the ${}^1S_0 - {}^3P_1$ resonance. Each data point corresponds to a steady state situation for the particular laser detuning (acquisition time for each individual data point is 500 ms). With a change of the detuning of the cooling laser the kinetic energy and, as a consequence, the amplitude of motion, the strengths of the sidebands, and the level of fluorescence are modified. With a Lamb-Dicke parameter $k\sqrt{\hbar/2m\omega}$ for the secular motion of about 0.2, a vibrational excitation as low as $\langle n \rangle = 3$, corresponding to a temperature of about 0.2 mK, already produces first order sidebands with a strength of 10% of the carrier.

To obtain the spectrum shown in Fig. 2(a) electric stray fields in the trap were compensated carefully by applying dc voltages to additional electrodes mounted close to the trap. This ensured that the ion was located in the center of the oscillating quadrupole potential so that no additional micromotion sidebands appear. At zero detuning the carrier of the spectrum can be seen with an asymmetric line profile. For small positive detunings a sharp drop in the fluorescence intensity is observed because at this point the ion is heated strongly through nonresonant excitations of blue secular sidebands. At the ensuing high vibration amplitudes, the ion stays in the trap but the laser excitation becomes so inefficient that fluorescence can hardly be detected above the stray light background. The low-frequency wing of the carrier shows essentially a saturation broadened Lorentzian line shape, as expected for an ion at rest. The sidebands of the secular motion at low frequencies contribute only little to the scattering rate, indicating that the ion is cold and well localized, fulfilling the Lamb-Dicke criterion.

In order to enhance the additional micromotion sidebands and to enlarge the capture range for sideband cooling at blue detuning the ion was shifted out of the trap center by applying a static field. The amplitude of this micromotion is fixed by the ion's displacement off the trap center and, in contrast to the secular motion and the ordinary micromotion, is independent of the laser detuning. The modulation index of the additional micromotion of the absorption spectrum in Fig. 2(b) is chosen to be ~ 2 (as can be seen from the left part of the spectrum where $\delta < 0$).

In the spectrum shown in Fig. 2(b), in addition to the carrier prominent sideband structures are visible, characterized by regions of elevated fluorescence. These regions start at negative detunings that are multiples of the trap frequency Ω (in the figure at -30, -20, and -10 MHz, respectively) and break down at about the midpoint between these frequencies (in the figure at -25 , -15 , and -5 MHz). This kind of structure is a signature of heating processes at negative detuning and has been analyzed in detail in Ref. [17]. For laser detunings below the additional micromotion sidebands $l\Omega$, cooling is very efficient since the laser detuning is negative with respect to the micromotion sideband as well as

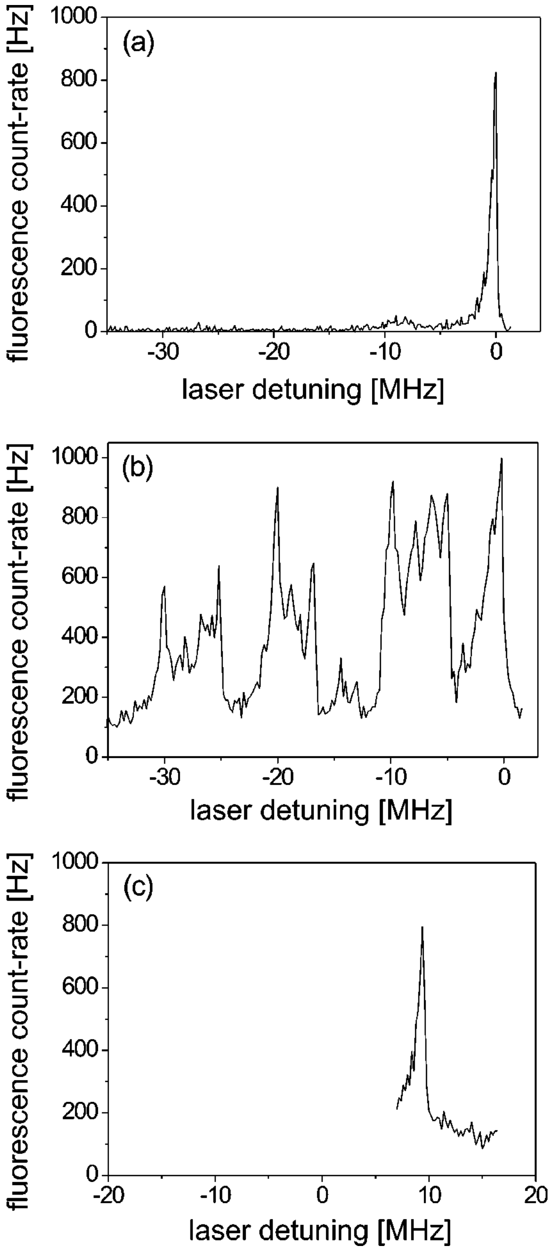

FIG. 2. Excitation spectra of a single trapped In^+ ion as a function of laser detuning. (a) The ion is localized in the center of the trap; no additional micromotion sidebands are visible. (b) Due to electric dc fields the ion is shifted off the center of the trap leading to sidebands of the additional micromotion at $\delta = l\Omega$, with $l=-1, -2, -3$ ($\Omega/2\pi = 10.03$ MHz). (c) After jumping with the laser detuning from $\delta/2\pi = -3$ MHz to $+7$ MHz, a similar line profile at $+10$ MHz as in (a) at zero detuning is obtained. The observation of this narrow resonance is the experimental proof for cooling at positive detuning (note the shift of the frequency scale).

with respect to the carrier. Here, the particle is cooled toward the motional ground state of the trap so that the sidebands of the secular motion are only weakly excited. As a result, a narrow Lorentzian half profile is obtained, analogous to the carrier. The regions of elevated fluorescence which appear for detunings on the blue side of the micromotion sidebands $l\Omega$, $l=-1,-2,-3$, correspond to higher excitation of the

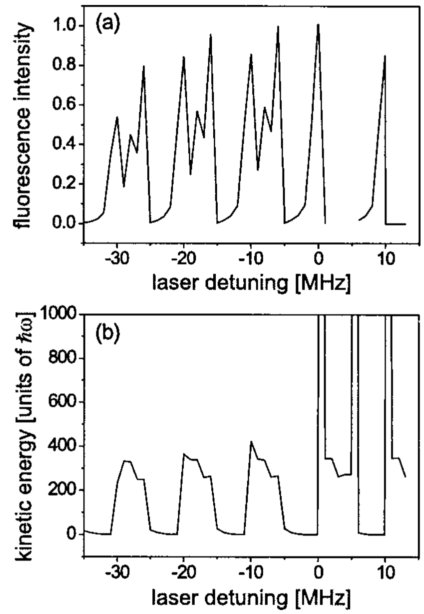

FIG. 3. Numerical calculation of (a) fluorescence and (b) mean kinetic energy as a function of laser detuning according to the semiclassical model of sideband cooling in a Paul trap. The fluorescence intensity is normalized to the emission of the ion at resonance; parameters correspond to those of Figs. 2(b) and 2(c).

secular motion. Here, the positive detuning with respect to the micromotion sidebands $l\Omega$ gives rise to heating but the negative detuning with respect to the carrier is a contribution to cooling. In a steady state both contributions are balanced. This happens at an elevated amplitude of the secular motion (corresponding to vibrational energies of a few hundred $\hbar\omega$) and leads to the observed increase of the fluorescence rate.

Under the conditions of the experiment shown in Fig. 2(b) cooling with positive detuning was attempted close to the first blue micromotion sideband at $\delta/2\pi = +10$ MHz. To prepare the ion at a temperature sufficiently low to be within the first capture range shown in Fig. 1, cooling to a steady state was obtained at a negative detuning -3 MHz. The laser detuning was then switched abruptly to a positive value of $+7$ MHz and continuous scanning toward higher positive detuning was started from that point. Switching the laser frequency is necessary since for smaller positive detunings heating processes dominate [see Fig. 3(b)]. The excitation spectrum obtained with this procedure is presented in Fig. 2(c): It exhibits a narrow Lorentzian half profile, similar to that shown by the cold ion on the carrier [Fig. 2(a)], but centered at the positive detuning of 10 MHz, indicating efficient cooling to low vibrational energy.

The experimental result can be well explained within our semiclassical model: On the red side of the $l=+1$ additional micromotion sideband, i.e., for detunings $+6\ \mathrm{MHz} \leqslant \delta/2\pi < 10$ MHz, cooling the ion is more efficient than heating. This is because heating via the blue secular sidebands originating from the carrier is less efficient than cooling via the red secular sidebands originating from the additional micromotion sideband at $+\Omega$.

For a quantitative comparison, Fig. 3 shows the fluorescence intensity and the kinetic energy of the energetically lowest stable steady state for each value of the laser detuning, as calculated numerically from the semiclassical model. The experimental procedure of jumping into the first capture range at positive detuning corresponds in the simulations to setting the initial energy to a low value in the search for an equilibrium. For the range of positive detunings +6 MHz $\leqslant \delta/2\pi < +10$ MHz similar values of the steady state kinetic energy are obtained as for the detunings −4 MHz $\leqslant \delta/2\pi < 0$ MHz, demonstrating that efficient cooling with positive laser detuning is indeed obtained.

IV. CONCLUSION

In conclusion, we have experimentally demonstrated the feasibility of laser sideband cooling at positive detuning for an ion in a radiofrequency trap. This verifies predictions of two theoretical models treating the nonlinear interaction of an ion in a time-dependent potential with monochromatic light. While this cooling method does not allow one to reach lower temperatures than sideband cooling with negative detuning and while it has only a limited capture range, it may nevertheless lead to an enhanced flexibility in the choice of experimental parameters. The method uses the micromotion in a radiofrequency trap, which is especially important in the case of trapped Coulomb crystals of several ions. It may be possible, for example, to cool two different isotopes with different optical frequencies simultaneously with only a single laser frequency, if this is tuned to the average frequency of the isotopes and the trap frequency is chosen as a subharmonic of the isotope shift.

[1] See, for example, J. Opt. Soc. Am. B **20**, 883 (2003), special issue.

[2] The notable exception from this statement is subrecoil laser cooling, based on a random walk in momentum space and the accumulation of atoms in a nonabsorbing state of well defined momentum. See F. Bardou, J. Ph. Bouchaud, A. Aspect, and C. Cohen-Tannoudji, *Levy Statistics and Laser Cooling* (Cambridge University Press, Cambridge, U.K., 2002).

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