# Doppler Cooling Trapped Ions with a UV Frequency Comb

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We demonstrate Doppler cooling of trapped magnesium ions using a frequency comb at 280 nm obtained from a frequency tripled Ti:sapphire laser. A comb line cools on the $3s_{1/2} - 3p_{3/2}$ transition, while the nearest blue-detuned comb line contributes negligible heating. We observe the cooling-heating transition and long-term cooling of ion chains with several sympathetically cooled ions. Spatial thermometry shows that the ion is cooled to near the Doppler limit. Doppler cooling with frequency combs has the potential to open many additional atomic species to laser cooling by reaching further into the vacuum and extreme ultraviolet via high-harmonic generation and by providing a broad bandwidth from which multiple excitation sidebands can be obtained.

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Doppler cooling with lasers was proposed in 1975 almost simultaneously for the cases of dilute atomic gases [1] and bound ions [2]. The first demonstrations followed in 1978 [3,4] and the technique has since become ubiquitous in atomic physics. Atom cooling is vital for precision measurements since it provides the means to overcome the limitations in precision spectroscopy imposed by the motion of the atoms, such as limited interaction times and Doppler broadening. In combination with the ability to trap, it is possible to attain virtually unlimited interaction times with, for example, a single ion at millikelvin or submillikelvin temperatures [5,6].

Traditionally, Doppler cooling is performed with spectrally narrow radiation obtained either directly or from nonlinear frequency conversion of infrared and visible continuous-wave lasers. While this approach has been exceptionally successful, it also sets severe restrictions on the accessible atomic species to those with strong cycling transitions in a wavelength range amenable to phase matching in nonlinear crystals, which is typically $\lambda > 190$ nm and, with more recently developed but not commercially available materials, $\lambda > 153$ nm [7]. Unfortunately, many atomic species of considerable interest have their lowest lying transitions in the vacuum ultraviolet (VUV), where the generation of continuous-wave laser radiation is extremely challenging [8,9]. To circumvent this problem, laser cooling using pulsed laser sources has been proposed [10]—in particular, with laser frequency combs, which have the double advantage of simultaneously providing the high peak powers to drive higher-order nonlinear optical processes and the long coherence of cw lasers. In this manner, it is possible to imagine a scenario where a frequency comb in the visible or near infrared is used to seed a high-harmonic generation stage to obtain coherent radiation in the vacuum and extreme ultraviolet [11], which is then used for cooling ions or atoms.

In brief, three forms of cooling with pulsed lasers can be identified [10]: (1) cooling with pulse trains with a repetition period longer than the upper-state lifetime of the atom, in which case the ion does not build upper-state population coherently across pulses and the cooling limit is only related to the inverse of the pulse duration [12], (2) cooling with a comb on a two-photon transition, where the small matrix element is partially compensated for by the fact that all of the comb lines contribute to the cooling in a pairwise fashion [10], and (3) cooling on a dipole-allowed transition using a pulse train with a repetition period shorter than the upper-state lifetime, in which case upper-state population is coherently built across many pulses such that the ion essentially interacts with a single comb line from the comb. In the latter case there is the additional possibility of using different comb lines to cool multiple species or, alternatively, to address far-detuned repumper transitions, as proposed in Ref. [13] and also demonstrated here.

In the proposal in Ref. [10], emphasis is made on cooling hydrogen on the two-photon $1s - 2s$ transition and carbon, which needs multiple far-detuned sidebands, all of which could, in principle, be provided by a single frequency comb. In addition to these examples it would also be advantageous to directly cool on certain dipole-allowed transitions in the VUV using a comb line from a frequency comb. For example, cooling a trapped Al ion on the 167 nm $3s^2 - 3s3p$ transition for the operation of the $Al^+$ optical atomic clock [14] or $He^+$ where a comb at 30 nm could be used to drive the $1s - 2p$ transition to cool it for the purpose of performing high-precision spectroscopy [15]. In this Letter we demonstrate cooling trapped ions on a dipole-allowed transition by using a single comb line from a frequency comb. To the best of our understanding, this is the first time that cooling has been demonstrated using the comb structure of a pulsed laser with a broadband spectrum. We hope that it provides the motivation for further experiments to be performed in the cooling of additional atomic species with frequency combs obtained from high-harmonic generation processes.

 

In Ref. [12] a mode-locked laser pulse train is used to cool trapped ions and in Ref. [16] the sixth harmonic of a pulsed dye laser is used to cool atomic hydrogen. In the latter experiment, the laser system runs at a very low repetition rate (50 Hz) without pulse-to-pulse phase stabilization. Therefore, no stable comb structure arises from the pulse train and the cooling process is essentially identical to that by cw laser. In the former case the pulses are short relative to the upper-state lifetime, but the comb line to comb line spacing is small compared to the atomic transition linewidth and the envelope of the laser spectrum is red detuned from the atomic resonance to provide cooling. Under this condition the cooling is not delivered by a narrow linewidth laser and, therefore, the minimum achievable temperature is much larger ($T_{\min} \approx \hbar\Delta\omega_{\text{Laser}}/2k_B$, as opposed to $T_{\min} \approx \hbar\Gamma_{\text{atom}}/2k_B$). In essence, the cooling is provided without utilizing the pulse-to-pulse coherence. On the contrary, when the comb line to comb line spacing becomes larger than the atomic linewidth, the photon scattering process happens across many pulses and the narrow linewidth of a single comb line can be accessed.

In the context of atomic beams, the possibility of cooling atoms into discrete velocity classes has been theoretically explored [17] and demonstrated [18]. The atoms thus cooled form, then, a series of sharp peaks at equidistant positions in velocity space. Another interesting demonstration using frequency combs to exert mechanical forces on cold atoms can be found in Ref. [19], where the use of interleaved pulse trains clearly shows that the mechanical force is a comb-related effect and that the upper-state population is coherently accumulated (or dissipated) across several pulses.

Our trapping apparatus [20] consists of a linear rf quadrupole trap driven at 22 MHz, with axial confinement provided by ring-shaped electrodes held at constant voltage. Micromotion compensation is provided by additional rods held at constant voltages. The radial and axial secular frequencies are $\omega_r \sim 2\pi \times 1$ MHz and $\omega_{\text{ax}} \sim 2\pi \times 45$ kHz, respectively. To load ions into the trap, neutral atomic Mg vapor of natural abundance is generated by heating a Mg wire in an atomic oven. A Rhodamine 6G laser pumped by the second harmonic of a $Q$-switched Nd:YAG laser is used to produce pulses at 570 nm, which are then frequency doubled to obtain $\mu$J-level pulses at 285 nm. This beam is used to resonantly photoionize neutral magnesium. For initial loading and cooling, a single-frequency dye (Rhodamine-19) laser at 560 nm is used after doubling in a bow tie enhancement cavity. $Mg^+$ is an alkalilike system with a single valence electron and ground state configuration $3s^1$. The $3s - 3p$ transitions at ~280 nm are dipole allowed and the excited state rapidly decays to the ground state, making these transitions ideal for Doppler cooling. A diagram of the relevant states is shown in Fig. 1(a). Both the $D_1$ and $D_2$ lines have ~3.8 ns excited-state lifetime (~41 MHz transition linewidth).

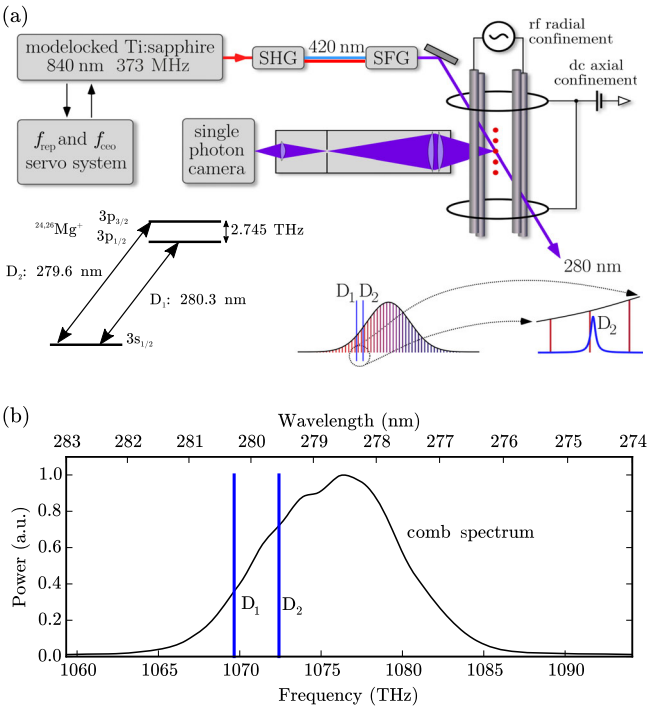


FIG. 1. (a) Schematic of the experimental setup. SPC: single photon camera, SHG: second-harmonic generation stage, SFG: sum-frequency generation stage and the comb spectrum after tripling into the UV. Blue lines, $D_1$ and $D_2$ lines of $Mg^+$. (b) Recorded comb spectrum. Vertical lines, $D_1$ and $D_2$ lines of $Mg^+$. Note that the envelope of the comb's spectrum is significantly blue detuned from both transitions.

The frequency comb is generated by frequency tripling a Ti:sapphire mode-locked ring laser operating at a repetition rate of 373 MHz with an output power of ~400 mW, a center wavelength of ~840 nm, and a spectral bandwidth (FWHM) of ~20 nm. The repetition rate is detected with a high-speed photodiode and stabilized via feedback to an intracavity piezoelectric-transducer-mounted mirror. Approximately 40% of the output power is coupled to a 16 cm-long piece of photonic crystal fiber and the octave-spanning broadened output is used in an $f - 2f$ interferometer to detect the carrier-envelope-offset frequency ($f_{\text{ceo}}$) [21], which is then mixed and stabilized feeding back to the pump power via an acousto-optic modulator. Both comb parameters ($f_{\text{rep}}$ and $f_{\text{ceo}}$) are locked to oscillators which are phase locked to a Global Positioning System–disciplined hydrogen maser, ensuring traceability of the frequency of each comb line. The remaining ~240 mW pulse train is first focused into a 1 mm thick $\beta$ barium borate (BBO) crystal to generate the second harmonic [22] at 420 nm. Further, the unconverted 840 nm pulse train is temporally overlapped with the 420 nm pulse train and focused in a second BBO crystal to generate the sum frequency at 280 nm. The 280 nm pulse train is sent through an acousto-optic modulator and the deflected beam with ~85% efficiency is focused into the trap. We typically

deliver 40–80 $\mu$W at the ions focused to a ~20 $\mu$m spot diameter. The overall efficiency of the third-harmonic process is only $3 \times 10^{-4}$, but we believe that the efficiency could be improved by fine-tuning the laser cavity to preferentially operate at the desired wavelength and by optimizing the nonlinear conversion stages. We estimate a power per comb line at the few nanowatt level, which provides an intensity on the order of 10 W/m$^2$, between 0.5% and 1% of the saturation intensity. At this comb line power we can readily observe the ion's fluorescence using a moderate (~3 s) integration time.

The fluorescence from the ions is collected by an $f/2$ four-lens condenser at a window close to the trap center and then imaged on the sensor plane of a microchannel plate single photon camera (Quantar Technologies, Mepsicron II) with a microscope objective. The entire imaging system provides a magnification of $\sim -100$. Given that all comb modes contribute to the background while only one produces ion fluorescence, spatial filtering of the ion's image is essential for observing spectral lines with a low background.

Because of the low power per comb line and the presence of multiple blue-detuned comb lines (in our case, the majority of them), we have not observed crystallization of hot ions using only the frequency comb. We believe that with several improvements this goal could be achieved in the near future. The steps to be taken would include a combination of (1) spectral filtering to shift the average wavelength to the red side of the transition, (2) increased average power to overcome the heating-cooling balance for initially hot ions which suffer from increased rf heating, and (3) using higher repetition rate combs to decrease the scattering from blue-detuned comb lines. As an additional resource, initial cooling using a buffer gas could be used. High repetition rate combs in the VUV and the extreme ultraviolet (XUV) via cavity-enhanced high-harmonic generation have been demonstrated [23], and their repetition rates could potentially be scaled further.

Once ions are loaded into the trap, we can switch to using the frequency comb for the last stages of cooling. We have observed that ions remain trapped and localized for extended periods of time while illuminated exclusively by the comb with a comb line in it red detuned from resonance. For example, Fig. 2(a) shows an image of a isotopically pure ($^{26}Mg^+$) and cold ion crystal kept for $> 5$ min while illuminated only with the frequency comb. It is important to note that the comb is tightly focused such that it mostly illuminates one ion and its intensity falls off towards the neighboring ions. We have chosen a tight focus to increase the scattering rate, which is crucial given the low power in our UV comb. It can be clearly seen that the comb is capable of keeping the entire crystal at low temperature, including the ions on the right side of the image, which are not significantly illuminated but remain cold due to the Coulomb interaction with the cooled ions. This effect is known as sympathetic cooling.

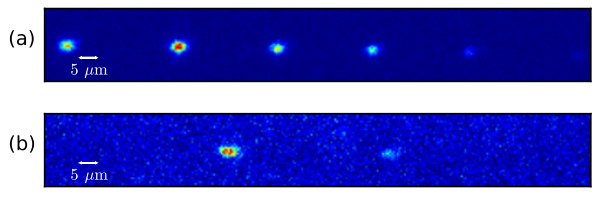


FIG. 2. (a) Image of a pure $^{26}Mg^+$ ion crystal integrated during 300 s exposure while illuminated only with the frequency comb. The ions remain clearly crystallized for the entire time. (b) Two $^{25}Mg^+$ ions cooled by the frequency comb. The scale bars on the images are 5 $\mu$m long.

The comb line to comb line spacing of the frequency comb is ~9 times larger than the ion's absorption linewidth ($\Gamma/2\pi = 42$ MHz, $f_{\mathrm{rep}} = 373$ MHz), which means that the scattering due to neighboring comb lines is reduced by a factor of $(\Gamma/2\pi f_{\mathrm{rep}})^2 \sim 10^{-2}$ and falls off quadratically with the mode-number difference. This also means that the excited-state population is coherently built across multiple pulses such that the comb structure is recovered as the comb is scanned across the resonance. To scan a comb line across the resonance, the repetition rate of the laser is changed in small increments. Given the large mode number involved ($N \sim 2.87 \times 10^6$), multiple comb lines can be scanned across the resonance with a mere $\Delta f_{\mathrm{rep}}/f_{\mathrm{rep}}$ of $\sim 10^{-6}$.

The ability to scan multiple comb lines across the resonance and directly collect fluorescence from the ions suggests a spectroscopy method to perform direct frequency comb spectroscopy on trapped ions. We have, in fact, employed this method to measure the involved transition frequencies and plan to publish the details elsewhere. Figure 3 shows the fluorescence collected while

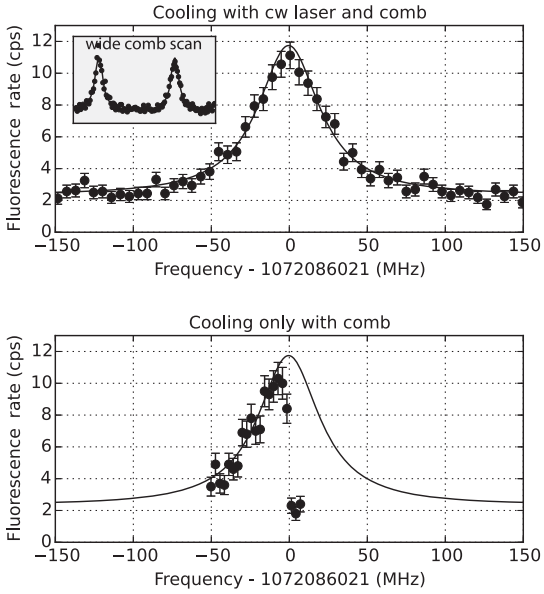


FIG. 3. Scan over line with and without additional cooling. The line in the lower plot is the fit to the spectroscopy data and is shown as a visual aid.

the repetition rate of the comb is changed such that a comb line is scanned across the $^{26}Mg^{+}$-$D_2$ transition. For spectroscopy runs, an additional cw laser is time interleaved to provide cooling and the ion's fluorescence is collected only during the periods when the comb is on. Conversely, to demonstrate cooling with the comb, we completely remove this additional cw laser and record fluorescence counts as the comb is scanned across the resonance. As the comb line is scanned across the resonance, it cools the ion while it is red detuned and starts heating the ion when it is very close to resonance and blue detuned. The heated ions acquire large trajectories and, therefore, do not interact with the laser anymore, causing a sharp drop in fluorescence counts. This cooling-heating transition leads to the observed asymmetric line shape in Fig. 3(b). It should be emphasized that the envelope of the comb's spectrum is significantly blue detuned from the transition frequency [see Fig. 1(b)] during the entire measurement, which clearly shows that the cooling effect is due to the comb structure under the envelope and, therefore, relies on the long pulse-to-pulse coherence.

Finally, to determine the equilibrium temperature of the ion, we perform a spatial thermometry measurement following the method in Ref. [24]. In brief, three ions are loaded into the trap and the axial secular frequency is measured at several ring voltages and images are recorded. For the case of three ions, the nearest-neighbor ion-to-ion distance is $[\kappa Z^2 e^2/(4\pi\epsilon_0 M\omega_{\mathrm{ax}}^2)]^{1/3}$, with $\kappa = 5/4$ [25]. Using the space to image size calibration, it is possible to accurately characterize the spread of the time-averaged ion's position in the more loosely confined direction. For a thermometry measurement as a function of laser detuning, we first reduce the axial confinement to $\omega_{\mathrm{ax}} \approx 2\pi \times 29$ kHz and then record several images at each detuning and fit a Gaussian shape to the ion's image on the axial direction. The thermal spread of the ion's position ($\Delta z_{\mathrm{thermal}}$) is recovered from the time-averaged image spread via the previously obtained calibration in combination with the known point-spread function of the imaging system. Finally, the ion temperature can be obtained from $\Delta z_{\mathrm{thermal}}$ through [24]

$$T \approx \frac{m\omega_{\mathrm{ax}}^2 \Delta z_{\mathrm{thermal}}^2}{k_B},$$

where $\omega_{\mathrm{ax}}$ is the axial secular frequency. This expression is only valid in the weak binding regime (i.e., the average number of phonons $\gg 1$).

Figure 4 shows the result of the thermometry measurement. The dashed line is a plot of the Doppler-limited temperature given by [5]

$$T = \frac{\hbar\Gamma}{8k_B}(1+\zeta)\left((1+s)\frac{\Gamma}{2|\Delta|} + \frac{2|\Delta|}{\Gamma}\right),$$

where $\Gamma$ is the ion's transition linewidth, $\Delta$ is the frequency detuning of the nearest comb line to the center of the transition, $s = I/I_{\mathrm{sat}}$ is the saturation parameter, and $\zeta = 2/5$ is a geometrical factor due to the dipole emission pattern. The dashed line in Fig. 4 is plotted using a saturation parameter similar to the experimental conditions ($s = 10^{-2}$). The data points show the measured ion temperatures at several detunings. At a detuning of $-0.45\Gamma$, we observe a temperature of 1.16(7) mK. The stated uncertainty and the error bars shown in the plot are of a statistical nature, arising from fitting a Gaussian profile to the ion image for each detuning. Other systematic effects such as residual micromotion induced by a possible mismatch between the axial and radial potential minima could result in overestimating our temperature measurement by as much as 0.3 mK (see Ref. [24] for a detailed analysis of the systematics).

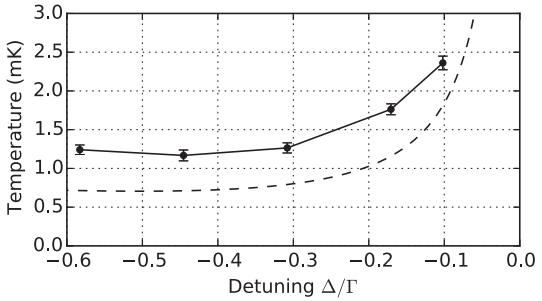


FIG. 4. Spatial thermometry measurement of a single ion while cooled with the frequency comb. The ion's temperature is measured by carefully quantifying its spatial extent in a trap with relaxed axial potential. The error bars indicate the statistical uncertainty from fitting a Gaussian profile to the ion image. The solid line joining the data points is shown as a guide for the eye. The dashed line indicates the Doppler-limited temperature as a function of laser detuning for a saturation parameter of $10^{-2}$.

Although the temperature limit in the equation above is only valid for cooling with a single line, we have calculated the effect of heating with a blue-detuned line for different comb line to comb line spacings and, in the case of $\sim 9 \times \Gamma$, the additional heating leads to a temperature increase of $< 10\ \mu$K over the entire range of our measurement. We expect that the effect of the remaining blue-detuned comb lines will be further diminished by the extremely low scattering rates, which fall off as the square of the mode-number difference. It should be noted that the achieved temperature is practically the same as that obtained when the ion is cooled with a cw laser and that the gap between the Doppler-limited and measured temperatures can be attributed to residual micromotion and other heating independent of the cooling method [24].

In additional experiments, we have also observed cooling of $^{25}Mg^{+}$, which has a hyperfine structure. When we use circularly polarized light and one comb line slightly red detuned from the $D_2$ $(F, m_F) = (3,3) \rightarrow (4,4)$ cycling transition, we observe that the ion is cooled, regardless of whether an additional comb line is tuned to the $D_1$ $(F = 2 \rightarrow F = 3)$ transition. More interestingly, with

linearly polarized light we observe that cooling is only possible for the case when one comb line is red detuned from the $D_2$ ($F = 3 \rightarrow F = 4$) and another comb line is close to resonance on the $D_1$ transition line ($F = 2 \rightarrow F = 3$), which is located $> 2.7$ THz away. We believe this to be clear evidence that the $D_1$ line works as a repumper to prevent population trapping in the ground state $F = 2$ levels. One could potentially also use the $D_2$ ($F = 2 \rightarrow F = 3$) by changing the repetition rate such that two comb modes match the hyperfine splitting. Nonetheless, using the $D_1$ line demonstrates an additional capability of the comb, which is that it can address far-detuned repumper transitions.

In conclusion, we have demonstrated for the first time Doppler cooling of trapped ions using a comb line from a frequency comb. We do not observe a degradation on the final temperature as compared to Doppler cooling with a cw laser. Finally, we also observe the repumping effect of a comb line exciting a transition $> 2.7$ THz away from the main cooling transition when cooling $^{25}Mg^{+}$.

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