Laser-induced forced evaporative cooling of molecular anions below 4 K

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The study of cold and controlled molecular ions has been of key importance for a wide range of applications, such as the production of cold antihydrogen, creation and study of anionic Coulomb crystals and in atmospheric research and astrochemistry. However, the commonly used anion cooling technique via collisions with a buffer gas is limited by the temperature of the used cryogenic cooling medium. Here we demonstrate the forced evaporative cooling of anions via a laser beam with photon energies far above the photodetachment threshold of the anion. We cool an anionic ensemble from an initial temperature of 370(12) K down to 2.2(8) K. This results in a three orders of magnitude increase in the phase-space density of the ions, approaching the near-strong Coulomb coupling regime. We present an analysis of the cooling dynamics through a thermodynamic model that includes the role of intrinsic collisional heating, without any fitting parameters. This technique can be used to cool any anionic species below liquid helium temperature, providing a tool to push the frontiers of anion cooling below state-of-the-art temperature regimes.

Recent technological developments in the hybrid trapping of ions and atoms have provided key insights into the interaction dynamics of molecular ions with atoms1. These studies involve investigations of the atom–ion reactive collisions2–8, as well as elastic and inelastic collisions, cooling the ions in their external (translational) and internal (vibration and rotational) degrees of freedom, respectively2–13. Although most of these systems primarily involve cationic ions, cooling anions to the cold—and even the ultracold—regime is of great interest in the study of astrochemically relevant reaction dynamics, that is, the growth of anionic carbon chains detected in Titan’s atmosphere14–16, associative detachment reactions of hydrogen in the early Universe17–19, or proton-exchange formation pathways of ammonia in interstellar media20,21. The outermost electron of a negative ion is bound by a short-range potential, which results in a lack of multiple stable electronic states, and due to their fragile nature, most anions cannot be cooled by traditional laser cooling methods. Although few promising candidates exist22–24, they still need precooling techniques, and direct Doppler cooling of anions has been experimentally elusive. Building on the success of evaporative cooling to create ultracold atoms, the forced evaporative cooling of anions was proposed three decades ago25. For a static laser-beam position, laser-induced evaporative cooling was demonstrated in atomic anions, where O− anions were cooled from 1.15 eV (13,000 K) to 0.33 eV (3,800 K) (ref. 26) in 100 s. Alternatively, the evaporative cooling of trapped antiprotons to temperatures as low as 9 K was achieved by altering the trap’s potential landscape27. In our work, we employ laser-induced evaporative cooling to cool molecular anions and demonstrate the generality of this technique. We reach temperatures below 4 K, which sets the limit of buffer-gas cooling using cryogenic helium. We remove the high-energetic tail of a molecular anionic ensemble, stored in a radio-frequency (rf) ion trap, via selective photodetachment. Subsequent rethermalization leads to efficient cooling of the remaining ions, which can be further enhanced by sweeping the detachment laser into the ion cloud (‘forced evaporative cooling’). This cooling process is well modelled by an ab initio thermodynamic calculation, revealing the role of rf heating induced by ion–ion collisions. With this approach, we cool down an ensemble of hydroxide (OH−) anions from 370(12) to 2.2(8) K.

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We achieve a Coulomb coupling parameter of $\Gamma = 0.06(5)$ in less than 4 s along with an increase in the ensemble's relative phase-space density by three orders of magnitude.

In our experiment, we trap a cloud of $\text{OH}^-$ anions in an rf octupole wire trap (Fig. 1a). A description of the experimental details is given in the Methods section. By addressing ions in a chosen axial interval, that is, via photodetachment with a focused laser beam, we can manipulate the ensemble's mean energy. To perform evaporative cooling, ions with a total energy higher than the mean energy must be removed. With this approach, one is able to preserve the trapping potential landscape, compared with the forced evaporative cooling of ions by lowering the trap depth. We measured the evolution of temperature and the number of ions for six laser-beam positions. The position and width of the laser beam compared with the ions' initial distribution is duly accounted for in determining the error. The measurement for the centre of the trap is limited by the accuracy of the translational stage used for these measurements approximated at 15 μm.

Fig. 1 | Experimental setup and thermalization dynamics under position-dependent photodetachment. a, Schematic of the octupole wire trap with hollow-core endcaps for the loading and extraction of ions. Perpendicular to the trap axial axis (into the plane of the paper), we shine a laser beam for photodetachment tomography and energy-selective evaporation. b, Axial ion distribution in the trap at 370(12) K. The solid points represent the photodetachment rates, which are proportional to the local column density. Each rate is determined by fitting an exponential decay to a loss measurement depending on the photodetachment time. The error bars show the ±1σ uncertainty of these fits. The solid curve is a fitted Gaussian function. Six different laser-beam positions and their widths are illustrated by the coloured vertical lines. The laser-beam position is centred along the vertical axis (x axis) and is moved along the axial direction (z axis). The laser-beam intensities (in units of watts per square centimetre) going from left to right are 15.1, 11.0, 3.8, 4.5, 11.7 and 20.0 (Supplementary Section I). The error bars represent the statistical errors derived from the decay measurements for each point. A decay measurement totalling 800 runs was recorded for each position with a mean of 35 detected ions. c, d. Temporal evolution of temperature (c) and the normalized ion number for the two laser-beam positions (d). The open circles represent the measured data, and the solid lines show the numerical integration of equations (1) and (2). The error bars represent the statistical error including the error propagation from the ion temperature determination. The measurements for each point were repeated until the width of the TOF of the ions converged to a certain value. This corresponds to a different sample size for each point, which is duly accounted for in determining the error. The measurement for the centre of the trap is limited by the accuracy of the translational stage used for these measurements approximated at 15 μm.

$T = \frac{3}{5} \left( -\frac{\sigma_{\text{pd}}}{k_B} \right) \int \rho_{\text{ion}}(x,y,z,T) \phi(x,y,z,\xi_1,\xi_2) V(x,y,z)^{\text{d}x\text{d}y\text{d}z} + \frac{5\sigma_{\text{pd}}}{6} T \int \rho_{\text{ion}}(x,y,z,T) \phi(x,y,z,\xi_1,\xi_2) \text{d}x\text{d}y\text{d}z$, \hspace{1cm} (1)

$\dot{N} = -\sigma_{\text{pd}} N \int \rho_{\text{ion}}(x,y,z,T) \phi(x,y,z,\xi_1,\xi_2) \text{d}x\text{d}y\text{d}z$. \hspace{1cm} (2)

Here $\sigma_{\text{pd}}$ is the absolute photodetachment cross section ($\sigma_{\text{pd}} = 8.5 \times 10^{-13}$ m$^2$ at 660 nm (1.88 eV) (ref. 29)), $\rho_{\text{ion}}$ is the normalized ion distribution and $\Phi(x,y,z,\xi_1,\xi_2)$ is the Gaussian-distributed photon flux at position ($\xi_1, \xi_2$). The total potential $V$ is defined as the sum of the harmonic axial potential created by the static endcaps and the time-averaged rf potential in the radial direction. In an octupole trap, this ponderomotive potential is proportional to $r\rho$, where $\rho$ denotes the radial distance from the trap centre. The first term in equation (1) corresponds to the mean potential energy of the ions addressed by the photodetachment beam and the second term represents the mean...

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The solid lines include the heating rate of equation (3). In order to apply the virial theorem. A detailed description of the model is found in the Methods section. For evaporative cooling, the ensemble’s thermalization rate must be faster compared with the induced loss rate. To ensure the validity of this condition, the laser power is kept well below the threshold for a complete model very accurately describes the change in both temperature and ion number. In the case of static evaporation (Fig. 1c, d), the relaxation of this integration are shown in Fig. 1c, d (solid lines), respectively, for each laser-beam configuration. Evidently, with the exception of one position represented via the green curve, the model accurately describes the dynamics. Compared with the other curves, the discrepancy in the green curve could be attributed to a lower ratio of the ion–ion thermalization rate to the ion loss rate at these particular settings (Supplementary Section I). The ensemble is cooled for sufficiently large distances of the laser beam from the cloud centre, reaching temperatures of around 200 K after 10 s, and removing about 90% of the molecules.

Enhanced cooling can be achieved by dynamically moving the laser towards the trap centre. The resulting ion dynamics are shown for four different laser-beam velocities (Fig. 2a, b). Here moving the laser beam at 1 mm s⁻¹ (red curve) to the trap centre, initially with 1,128(17) ions in the mean, the final ensemble temperature is 2.2(8) K with a mean of 18.3(5) ions remaining in the trap. This corresponds to a decrease in the phase-space density by three orders of magnitude.

The dashed lines in Fig. 2a, b are the numerical integration of equations (1) and (2) including a dynamically moving photodetachment laser beam. Although the thermodynamic model describes the observed temperature evolution very well, the measured number of ions substantially deviates from the predicted numbers for lower beam velocities. This can be explained by the influence of ion–ion collisions, which transfer the energy of the driving rf field to the secular motion of ions. The resulting heating rate can be approximated by the product of the ion–ion collision rate, described by the Chandrasekhar–Spitzer plasma self-collision rate ν_{ii}(T) (ref. 30) and the relative energy transfer per collision ε:

$$T = \frac{3}{7} ν_{ii}(T) ε^{1/3} \frac{3}{7} \frac{n e^4 \log \Lambda_i}{12 \epsilon^2 \sqrt{n m(k_B T)}} \epsilon^{1/3} T.$$  

The mean rf motion in an octupole trap is given by \(\langle E_{rf}\rangle = \frac{1}{2} k_B T\) and the relative energy change per collision (ε = 0.045(6)) is measured by a background heating measurement (Supplementary Section II). Here \(n\) is the ion density and \(\Lambda_i\) is the Coulomb logarithm, where \(\Lambda_i\) is the ratio between the Debye length and the classical distance of the closest approach.

Equations (1) and (2) are independent of the absolute ion number, but the ion–ion collision rate is not. The detection efficiency of our setup is, therefore, determined by an independent thermalization measurement (Supplementary Section I). The zero-fit-parameter models, including the heating rate, are shown in Fig. 2 (solid lines). The complete model very accurately describes the change in both temperature and ion number. In the case of static evaporative cooling (Fig. 1c, d), the influence of ion–ion collisional heating is found to be negligible.

The efficiency and pace of cooling our ensemble are limited by two timescales, namely, the evaporation rate and thermalization rate. The evaporation rate is proportional to the detachment rate and thus the photon flux. If we move the laser beam fast enough, the removal of high-energy ions, the cooling efficiency of the evaporation rate exceeds the thermalization rate, and the ions are only spilled...
without an increase in the phase-space density of the ensembles. If we move it too slowly (for example, for laser-beam velocities of 0.50 and 0.25 mm s⁻¹), rf heating via ion–ion collisions becomes more prominent and results in an additional ion loss. In our system, cooling with 1 mm s⁻¹ (Fig. 2, red curve) is more efficient since we have more ions left in the end. For 2 mm s⁻¹ (Fig. 2, purple curve), the less cold final temperature is a result of the evaporation of the colder ions. The thermalization rate of a one-component plasma is proportional to the product of the Coulomb logarithm and the phase-space density of the ensembles, for which we see a tremendous increase by three orders of magnitude.

A one-component plasma is characterized by the dimensionless Coulomb coupling constant Γ, which represents the nearest-neighbour Coulomb energy to the thermal energy:

$$\Gamma = \frac{e^2}{4\pi \epsilon_0 k_B T(r)}$$

(4)

where \(r\) is the mean interparticle distance (Wigner–Seitz radius). In a weakly coupled plasma (\(\Gamma \ll 1\)), Coulomb interactions can be treated as two-particle collisions perturbing the system, as done in equation (3). For \(\Gamma \gg 1\), the assumption breaks down and reveals phenomena such as liquid-like behaviour and a liquid–solid phase transition at \(\Gamma = 178\) (refs. 31,32). Figure 2c shows the Coulomb coupling constant corresponding to the measurement shown in Fig. 2a,b. For the maximum forced evaporative cooling efficiency (laser-beam velocity of 1 mm s⁻¹ (Fig. 2, red curve)), the coupling parameter increases initially from \(\Gamma = 0.35(4) \times 10^{-3}\) to 0.06(3). Based on the model, one could achieve runaway evaporative cooling in the system, and the coupling constant could be further increased by increasing the detachment rate beyond the rate currently achievable in our experimental setting (Supplementary Section III).

The evaporative cooling of the ions in our system, as in the case of atoms, can also be quantified via the phase-space density. If \(n\) is the ion density and \(\lambda_{DB}\) is the thermal de Broglie wavelength, the phase-space density \(D\) is calculated as

$$D = n\lambda_{DB}^3 = n \left(\frac{2\pi n^2}{mk_B T}\right)^{3/2}$$

(5)

Figure 2c (inset) presents the calculated phase-space density for each measured point and the thermodynamic model (solid lines) relative to the initial conditions. We see that for a laser-beam velocity of 1 mm s⁻¹ (Fig. 2, red curve), the relative phase-space density increases by three orders of magnitude, an important indicator of the efficient cooling of the ions.

For particle densities as low as that in our system, evaporative cooling only works due to the long-range interaction between the ionic particles, ensuring sufficiently fast rethermalization times. Due to the short-range interaction between the neutral atoms, the evaporative cooling of atoms at these low densities would not be feasible, as the rethermalization time would be too long.

Genuinely, evaporative cooling goes along with losses, which ultimately limits the achievable phase-space densities. This is in contrast to sympathetic cooling through, for example, helium buffer gas or laser-cooled ions, which is, in principle, lossless. In addition, reaching lower temperatures with evaporative cooling would also require a high-precision trap design to achieve electric trapping potentials of the same order as the thermal energy. Dynamically manipulating the trap’s potential landscape could be another important addition to overcoming the temperature limits of our current implementation of evaporative cooling.

Our experimental results in combination with the underlying predictive model of the ion dynamics show that forced evaporative cooling via above-threshold photodetachment is an efficient tool to reduce the thermal energy of, in principle, any anionic species by orders of magnitude. This establishes a way to translationally cool anions below the temperature of liquid helium without the need for a buffer gas, thus ensuring ultrahigh-vacuum conditions. This would allow entering the regime of low-energy scattering resonances, as has recently been achieved with atomic cations, eventually approaching the regime of strong Coulomb coupling. In a multispecies anionic plasma, this laser-induced cooling provides the advantage of addressing one anionic species and sympathetically cooling the other species, for example, antiprotons, without the loss of the latter. The cooling of molecular anions can even be extended to address internal degrees of freedom by tuning the laser-light frequency close to the anion’s photodetachment threshold.

**Online content**

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-023-02084-6.

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Methods

Experimental details
Our molecular anion of interest, the OH⁻ anion, is created in a pulsed plasma discharge source and subsequently trapped in an octopole wire trap. This trap combines the advantage of reduced rf heating in multipole traps and the use of wires to provide optical access to investigate atom–anion interactions in a hybrid trap.18–20. The measurements were performed on trapped ions initially thermalized with a room-temperature helium buffer gas, which result in an ensemble with a temperature slightly above the buffer gas. This increase in temperature is attributed to rf heating. We determine the temperature of ions in the trap by extracting them through a hollow-core endcap and subsequently detecting the time of flight (TOF) for each particle on a microchannel plate. With an optimized extraction potential, one can linearly map the energy distribution onto the TOF to the detector. A detailed description of the setup and TOF thermometry can be found elsewhere.

Although the energy distribution is obtained from the TOF, the spatial distribution of the ions in the trap can be measured via above-threshold photodetachment tomography.11. In our setup, the laser light with a photon energy of 1.88 eV, which is above the photodetachment threshold of OH⁻ (1.83 eV) (ref. 42), is focused down to a diameter of 180 μm (1/e²) and can be moved by a translational stage. One measurement of the ion distribution along the trap axis, after initialization with the helium buffer gas pulse, is shown in Fig. 1b (solid points). Since the axial distribution is well described by a harmonic potential, a Gaussian distribution of the ion cloud can be observed.

Thermodynamic model
The approach of this rate model is based on another work, and it assumes that the cooling process is slow enough such that the ions are in thermal equilibrium at any time. This way, the total energy can be described by the sum of the mean kinetic energy and mean potential energy:

\[ \mathcal{E}_{\text{tot}} = \langle \mathcal{E}_k \rangle_y + \langle \mathcal{E}_p \rangle_y = \frac{3}{2} k_b T + \frac{5}{6} k_b T = \frac{7}{3} k_b T. \]

(6)

The prefactor \( \frac{7}{6} \) is the result of the virial theorem for an octopole trap. The change in ion number is described by the overlap integral:

\[ \mathcal{N} = -N \langle \sigma \phi \rangle_y = -N \int \sigma(v) \phi(r) \rho_{\text{ion}}(r, v) dr dv \]

(7)

Here \( \sigma \) is the absolute photodetachment cross section, \( \rho_{\text{ion}} \) is the normalized ion density and \( \phi \) is the photon flux. The change in total energy is given by the energy of the lost ions:

\[ \langle \mathcal{E}_\text{tot} \rangle_y = -N \langle \sigma \phi \mathcal{E}_p \rangle_y \]

(8)

\[ \Rightarrow \langle \mathcal{E}_p \rangle_y = -\langle \sigma \phi \mathcal{E}_p \rangle_y + \langle \sigma \mathcal{E}_p \phi \rangle_y \]

(9)

\[ \Rightarrow -\langle \sigma \phi \mathcal{E}_k + \mathcal{E}_p \rangle_y + \langle \sigma \mathcal{E}_k \phi \rangle_y + \langle \sigma \phi \mathcal{E}_p \rangle_y = -\langle \sigma \phi \mathcal{E}_k + \mathcal{E}_p \rangle_y + \langle \sigma \mathcal{E}_k \phi \rangle_y + \langle \sigma \phi \mathcal{E}_p \rangle_y \]

(10)

Only the laser intensity and potential energy depend on \( r \) and the kinetic energy depends on \( v \). In the case of above-threshold photodetachment, the cross section is independent of the ion velocity. Thus, the kinetic-energy terms cancel out.

\[ \langle \mathcal{E}_p \rangle_y = -\sigma \langle \phi \mathcal{E}_p \rangle_y + \sigma \langle \mathcal{E}_p \phi \rangle_y \]

(11)

\[ \Rightarrow \frac{7}{8} k_b T = -\sigma \int \rho_{\text{ion}}(x, y, z, T) \phi(x, z, x_1, z_1) V(x, y, z) dx dy dz + \frac{5}{8} k_b T \sigma \int \rho_{\text{ion}}(x, y, z, T) \phi(x, z, x_1, z_1) dx dy dz \]

(12)

\[ \mathcal{N} = -\sigma N \int \rho_{\text{ion}}(x, y, z, T) \phi(x, z, x_1, z_1) dx dy dz - k_{\text{bg}} \mathcal{N} \]

(13)

\[ V(x, y, z) = 2.04 \times 10^{-11} \text{ m}^2 \times z^2 + 3.9(6) \times 10^{-10} \text{ m}^6 \times (x^2 + y^2)^3 \]

(14)

In general, forced evaporative cooling can be implemented for arbitrary starting conditions. For a known ion density distribution and photon flux, this model can be adapted to calculate the dynamics under any initial conditions.

Data availability
The data that support the findings of this study are available from the corresponding author upon request. Source data are provided with this paper.

Code availability
The codes and analysis files that support the findings of this study are available from the corresponding author upon request.

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Author contributions
J.T. and S.Z.H. performed the experiments and data analysis. J.T. simulated the thermodynamical model. M.N. provided the simulations on temperature determination. E.S.E., R.W. and M.W. conceptualized the experiments. J.T. and S.Z.H. wrote the initial draft. All the authors edited and contributed in the preparation of the final manuscript.

Competing interests
The authors declare no competing interests.

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