Realization of collective strong coupling with ion Coulomb crystals in an optical cavity

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Cavity quantum electrodynamics (CQED) focuses on understanding the interactions between matter and the electromagnetic field in cavities at the quantum level^{1,2}. In the past years, CQED has attracted attention^{3–9} especially owing to its importance for the field of quantum information¹⁰. At present, photons are the best carriers of quantum information between physically separated sites^{11,12} and quantum-information processing using stationary qubits¹⁰ is most promising, with the furthest advances having been made with trapped ions^{13–15}. The implementation of complex quantum-information-processing networks^{11,12} hence requires devices to efficiently couple photons and stationary qubits. Here, we present the first CQED experiments demonstrating that the collective strong-coupling regime² can be reached in the interaction between a solid in the form of an ion Coulomb crystal¹⁶ and an optical field. The obtained coherence times are in the millisecond range and indicate that Coulomb crystals positioned inside optical cavities are promising for realizing a variety of quantum-information devices, including quantum repeaters¹² and quantum memories for light^{17,18}. Moreover, cavity optomechanics¹⁹ using Coulomb crystals might enable the exploration of similar phenomena investigated using more traditional solids, such as micro-mechanical oscillators²⁰.

The strong-coupling regime of cavity quantum electrodynamics for a single two-level quantum system is reached when the coupling rate g at which a single photon is absorbed or coherently emitted into the cavity mode by the quantum system is larger than both the field decay rate κ out of the cavity and the dipole decay rate γ of the quantum system^{1,2}. Under these conditions, the quantum processes of absorption and stimulated emission dominate the dynamics and can lead to a coherent exchange of a photon in the cavity and an excitation of the quantum system. This regime has been successfully accessed with neutral atoms^{2,4,5}, and recently with quantum dots⁶ and Cooper pairs⁷. Although single ions have also been applied in CQED-related experiments^{3,21,22}, the single-particle strong-coupling regime has not yet been reached. For an ensemble of N quantum systems interacting with a photon in a specific cavity mode, a collective coupling rate $g_N \equiv gN^{1/2}$ is obtained. When g_N exceeds both κ and γ , the so-called collective strong-coupling regime is reached, which has so far only been realized using cold gases of neutral atoms in the form of atomic beams²³, magneto-optical traps²⁴ or Bose–Einstein condensates^{8,9}.

In contrast to neutral atomic gases, ion Coulomb crystals have a series of attractive solid-state properties of importance for collective strong-coupling CQED studies and for the development of quantum-information devices. In Coulomb crystals, the ions have a uniform density and are positioned at distances of $\sim 10 \mu\text{m}$ (refs 16, 25). Consequently, there are no close-distance interactions between the particles that would lead to either temporal or

spatial inhomogeneous spectral broadening. Ion Coulomb crystals can, furthermore, be stably trapped for hours, which allows for repeatable experiments with a coupling strength being constant to within a few per cent. Moreover, the crystal structure allows for various cavity modes to interact with the same crystal without interference and with identical coupling strengths. Finally, the low density of Coulomb crystals has the advantage that all of the interacting ions can very efficiently be optically pumped into a specific desired state.

To reach the collective strong-coupling regime, here we use a linear radiofrequency ion trap that incorporates an 11.8-mm-long optical cavity with its axis coinciding with the central trap axis²⁶ (z axis), as shown in Fig. 1a. With a moderately high finesse $F = 3,000 \pm 200$ at the 866 nm resonant wavelength of the $3d^2D_{3/2} - 4p^2P_{1/2}$ transition in $^{40}\text{Ca}^+$ addressed in the experiments, the cavity has a decay rate of $\kappa = 2\pi \times 2.15 \text{ MHz}$. The natural dipole decay rate of an excitation on the $3d^2D_{3/2} - 4p^2P_{1/2}$ transition is $\gamma = 2\pi \times 11.15 \text{ MHz}$. With the 5 mm focal length of both curved cavity mirrors, this leads to a single-photon coupling rate of $g = 2\pi \times 0.53 \text{ MHz}$ for an ion positioned at an anti-node of the standing-wave field in the centre of the TEM_{00} cavity mode. With these parameters, the collective strong-coupling regime is entered when the effective number of ions interacting with the photons in a cavity mode exceeds ~ 500 (see the Methods section).

The trap can be loaded controllably *in situ* with a few to tens of thousands of $^{40}\text{Ca}^+$ ions through isotope-selective two-photon ionization²⁶. The trapped ions are Doppler laser cooled into a Coulomb crystal^{16,25} by driving the $4s^2S_{1/2} - 4p^2P_{1/2}$ transition using two counter-propagating 397 nm light beams with left- and right-handed circular polarizations and by repumping on the $3d^2D_{3/2} - 4p^2P_{1/2}$ transition using an 866 nm light beam, propagating along the y axis and linearly polarized along x (Fig. 1a, b). Using a charge-coupled device (CCD) camera, projection images of the crystals can be recorded by collecting 397 nm fluorescence light emitted by the ions during the cooling process. From these images, the effective number of ions N in the crystal interacting with the TEM_{00} mode of the cavity can be extracted (see the Methods section). Figure 1c shows a projection image of a Coulomb crystal containing $6,400 \pm 200$ ions of which 536 ± 18 ions effectively interact with the cavity mode. For a visual impression of the overlap of the ions in the crystal and the TEM_{00} mode of the cavity, the repumping beam can be resonantly injected into the cavity. A resulting image is shown in Fig. 1d; in this case, the ions outside the cavity mode appear dark owing to the lack of repumping from the metastable $3d^2D_{3/2}$ -level. After the Coulomb crystallization and in the presence of a magnetic bias field along the z axis $\mathbf{B} = (B_x = 0.0 \text{ G}, B_y = 0.0 \text{ G}, B_z = 2.6 \text{ G})$, the ions are optically pumped with high efficiency ($\sim 97\%$) into the $m_j = +3/2$ Zeeman substate of the long-lived metastable

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Figure 1 | Schematic diagram and ion Coulomb crystal images. **a**, Schematic diagram of the experimental set-up with the linear radiofrequency ion trap incorporating an optical cavity along its radiofrequency-field-free axis (z axis). The incoupling mirror of the cavity is mounted on a piezoelectric transducer (PZT) allowing for tuning of the cavity around the atomic resonance. LC: laser cooling beam, RP: repumping beam, OP: optical pumping beam, PP: probe pulse, RB: reference beam, CM: cavity mirrors, PZT: piezoelectric transducer, CCD: CCD camera, APD: avalanche photodiode. **b**, Energy levels of $^{40}\text{Ca}^+$ including the relevant transitions for the three main parts of the experimental sequence. First, the ions are Doppler laser cooled by driving the $4s^2S_{1/2}-4p^2P_{1/2}$ transition using 397 nm laser beams and repumping on the $3d^2D_{3/2}-4p^2P_{1/2}$ transition by light at 866 nm. Next, the ions are optically pumped into the $m_j = +3/2$ Zeeman substate of the $3d^2D_{3/2}$ level using the optical pumping beam in combination with the laser cooling beams. Finally, the coupling of the Coulomb crystals to the cavity field is measured by injecting a weak probe pulse of 866 nm light into the cavity and detecting the reflection signal by an avalanche photodiode. **c,d**, Projection images of a 1-mm-long ion Coulomb crystal recorded with the CCD camera by collecting 397 nm fluorescence light emitted during laser cooling: the whole crystal (**c**) and ions within the cavity mode volume (**d**) (see the text for details).

$3d^2D_{3/2}$ -level of $^{40}\text{Ca}^+$ (lifetime: $\sim 1\text{s}$) using a beam making an angle of 45° with respect to the z axis and having a proper elliptical polarization (Fig. 1a,b).

To investigate the collective coupling of the ions in the Coulomb crystal to the TEM_{00} mode of the cavity, a 1.4- μs -long and 99%-mode-matched probe pulse is injected. The field strength of this pulse corresponds to an average intra-cavity photon number of about one or less at any time. The frequency of the probe light can be tuned around the $3d^2D_{3/2}-4p^2P_{1/2}$ transition, and it is left-handed circularly polarized to obtain the strongest coupling with the optically pumped ions (Fig. 1b). A weak off-resonant continuous-wave laser beam at 894 nm is also

injected into the cavity as a reference of the cavity fluctuations and drifts. Photons in the probe pulse and the reference beam are detected in reflection and transmission, respectively, by avalanche photodiodes with an overall efficiency of 16% after spectral and spatial filtering (Fig. 1a).

An accurate way to quantify the strength of the collective coupling is, for a series of detunings of the probe frequency ω_p with respect to the $3d^2D_{3/2}-4p^2P_{1/2}$ transition frequency ω_a , to measure the width and resonance phase shift of the reflected probe signal when scanning the cavity resonance frequency ω_c around ω_p . Figure 2a, b shows results of such measurements for a crystal similar to the one shown in Fig. 1. According to the two-level atom

Figure 2 | Collective coupling measurements. For all of the data presented, a crystal similar to that of Fig. 1c with 520 ± 18 ions effectively interacting with the fundamental TEM_{00} cavity mode was used. **a**, Measured half-widths of the probe signal, κ' , when scanning the cavity resonance frequency, ω_c , for a series of detunings of the probe from atomic resonance, Δ . The inset shows a sample set of 100 averaged scans of the cavity resonance frequency at a given frequency of the probe pulse ($\Delta = 10$ MHz). The widths $2\kappa'$ obtained from a Lorentzian fit of five of these data sets are averaged to give one point in the absorption curve (orange solid circle). **b**, The corresponding shift of the cavity resonance, δ'_c , as a function of Δ . The solid lines are fits to the theoretical absorption and dispersion curves (see the text for details). In **a** and **b**, the cavity resonance frequency is scanned over 1.3 GHz around the atomic resonance, ω_a , at a rate of 30 Hz. During the frequency scan, the cooling, optical pumping and probing sequence is repeated at a rate of 50 kHz, with a 1.4 μs probe detection time. **c**, Measured probe reflection spectra when locking the cavity resonance to the atomic resonance ($\omega_c = \omega_a$) and scanning the probe field frequency ω_p around the common cavity-atom resonance. The red data points (squares) are obtained with an empty cavity, whereas the blue data points (circles) are obtained with an ion Coulomb crystal present. The solid lines are theoretical fits²³. For each detuning of the probe from the common cavity and atomic resonance ($\omega_c = \omega_a$), one data point is the average of 20,000 sequences. The error bars are ± 1 s.d.

Figure 3 | Collective coupling dependence on the number of ions.

Cooperativity, C , versus the effective number of ions, N . The blue data points (circles) represent results obtained when applying left-handed polarized probe light, resonantly interacting with the ions, whereas the red data points (squares) are obtained with a right-handed circularly polarized probe where no coupling to the ions is expected to be observed. The solid lines are linear fits. The dashed horizontal line indicates the region above which collective strong coupling is achieved ($g_N > \kappa, \gamma$). The inset shows the coupling rate for a single crystal over 2 h. A constant coupling fit to the data (solid blue line) results in $C = 2.04 \pm 0.03$. All vertical error bars: ± 1 s.d., horizontal error bars: see the Methods section.

theory², the half-width of the probe signal κ' (Fig. 2a) should have the following Lorentzian dependence,

$$\kappa' = \kappa + g_N^2 \gamma' / (\gamma'^2 + \Delta^2)$$

where $\Delta = \omega_p - \omega_a$ is the probe detuning from the atomic resonance and γ' is the effective dipole decay rate. The corresponding cavity resonance phase shift δ'_c (Fig. 2b) should vary dispersively according to

$$\delta'_c = g_N^2 \Delta / (\gamma'^2 + \Delta^2)$$

A fit to the data of Fig. 2a with g_N and γ' as free parameters results in a collective coupling rate $g_N = 2\pi \times (12.2 \pm 0.2)$ MHz and a dipole decay rate $\gamma' = 2\pi \times (11.9 \pm 0.4)$ MHz. The value of g_N is in very good agreement with the calculated collective coupling rate of $2\pi \times (12.1 \pm 0.3)$ MHz using the effective number of ions $N = 536 \pm 18$ deduced from the CCD image and the optical pumping efficiency into the $^2\text{D}_{3/2}$ $m_1 = +3/2$ Zeeman substate of 97%. The slightly higher value of γ' as compared with the natural dipole decay rate $\gamma = 2\pi \times 11.15$ MHz is consistent with a Doppler broadening corresponding to a temperature of the crystal of 20–30 mK. Micromotion of the ions induced by the trapping radiofrequency field^{25,27} could potentially contribute to the observed broadening. However, both simulations of Coulomb crystals in linear ion traps²⁷ and observed identical coupling rates for the TEM_{00} and $\text{TEM}_{01,10}$ modes do not suggest measurable effects. A complementary method for quantifying the coupling strength is based on locking the cavity resonance to the atomic resonance ($\omega_c = \omega_a$) and scanning the probe field frequency ω_p around the common cavity-atom resonance. This leads to the observation of the so-called single-photon Rabi splitting arising from the resonantly coupled crystal-cavity system²³. From the

Figure 4 | Collective Zeeman substate coherence time measurements.

a, The blue points represent the relative change in the cooperativity, $(C(\tau) - \bar{C})/\bar{C}$, as a function of the delay time τ between optical pumping and probing, in the presence of a magnetic bias field having components both along the x axis and the z axis $\mathbf{B} = (B_x = 0.15 \text{ G}, B_y = 0.0 \text{ G}, B_z = 0.15 \text{ G})$. $\bar{C}$ is the mean cooperativity averaged over one oscillation period of the Larmor precession between the four Zeeman substates of the $3d^2D_{3/2}$ -level. A fit to the results (solid line) corresponds to a coherence time of $\tau_E = 1.7^{+100}_{-0.8} \text{ ms}$ or $\tau_G = 0.5^{+0.6}_{-0.2} \text{ ms}$ (see the Methods section). **b**, For comparison, the cooperativity values (normalized to the mean value of all points, C_m) for a bias field oriented along the z axis ($B_z = 0.15 \text{ G}$) are presented as well (red points). In all measurements, the effective number of ions interacting with the cavity field was $N = 570 \pm 20$. Error bars are $\pm 1 \text{ s.d.}$

measured Rabi splitting in the data presented in Fig. 2c, a collective coupling rate of $2\pi \times (12.2 \pm 0.2) \text{ MHz}$ is deduced, consistent with the previous results presented in Fig. 2a,b.

The effective number of ions N in the cavity mode volume can be controlled with high accuracy by varying either the crystal shape or density through changes of the trapping potentials. An example of the observed collective coupling effect dependence on N is presented in Fig. 3, where the cooperativity parameter $C = g_N^2/2\kappa\gamma'$ is plotted as a function of N . In accordance with the expression for g_N , the cooperativity scales linearly with N . For the largest number of ions in the cavity mode, $N \sim 1,600$, which corresponds to a Coulomb crystal length of 3 mm and an ion density of $6 \times 10^8 \text{ cm}^{-3}$, one obtains a cooperativity of 8, or an equivalent effective optical depth of 16. This shows that the collective strong-coupling regime can be entered with large ion Coulomb crystals, and the cooperativity supersedes previously obtained values with ions by at least an order of magnitude^{3,21,22}. Furthermore, the measured stable coupling strength for a single crystal over two hours presented in the inset of Fig. 3 highlights the solid-state nature of Coulomb crystals.

To evaluate the prospect of using ion Coulomb crystals for quantum storage, the temporal stability of collective Zeeman substate coherences in the $3d^2D_{3/2}$ -level is measured. In these experiments, the optical pumping light beam is propagating along the y axis and it is polarized linearly along the z axis and the bias magnetic field has components both along the x axis and the z axis $\mathbf{B} = (B_x = 0.15 \text{ G}, B_y = 0.0 \text{ G}, B_z = 0.15 \text{ G})$. After the optical pumping preparation, this bias field configuration gives rise to an induced coherent Larmor precession between the Zeeman substates of the $3d^2D_{3/2}$ -level with respect to the quantization axis defined by the propagation direction of the probe light (z axis). This coherent dynamics is probed by varying the delay between the end of the optical pumping period and the injection of the

probe pulse. The decay of the resulting coherent oscillations in the cooperativity presented in Fig. 4 leads to a Zeeman substate coherence time $\tau_E = 1.7^{+100}_{-0.8} \text{ ms}$ or $\tau_G = 0.5^{+0.6}_{-0.2} \text{ ms}$, assuming an exponential or Gaussian decay, respectively (see the Methods section). This value, obtained without any advanced shielding against stray magnetic fields, is already at the level of the best reported coherence times for single ions in equivalent magnetic-field-sensitive states²⁸.

The effective optical depth of 16 is comparable to those typically used in light storage experiments with neutral atoms¹⁸, and it is large enough for quantum state storage and retrieval with potential fidelities $>90\%$ (ref. 29). By conducting similar CQED experiments using two-component Coulomb crystals³⁰, it should be feasible in the near future to increase the optical depth by at least a factor of two, partly by confining more ions within the TEM_{00} mode volume, and partly by forcing the long-range ordered ions in the central crystal component³⁰ to 'line-up' with the anti-nodes of the standing-wave cavity field. Possibilities for multi-mode light storage can also be investigated. A further interesting aspect of CQED experiments with Coulomb crystals yet to be explored is the extent to which these observed collective effects may be used to reveal information about the crystals, such as structural phase transitions. Finally, CQED experiments with Coulomb crystals might provide insight into light-induced mechanical effects on more traditional solids such as micro-mechanical oscillators²⁰.

Methods

Determination of the effective number of ions. For an ensemble of N_{tot} ions, one can define the effective number of ions, N , interacting with a TEM mode of the cavity through the expression for the collective coupling strength $g_N \equiv gN^{1/2}$ with $N \equiv \sum_{i=1}^{N_{\text{tot}}} \psi^2(\mathbf{r}_i)$. Here, $\mathbf{r}_i$ is the position of the i th ion and $\psi(\mathbf{r})$ is the standing-wave mode function. For the considered TEM_{00} mode, one finds

$$\psi^2(\mathbf{r}) = \left(\frac{w_0}{w(z)} \right)^2 \exp(-2(x^2 + y^2)/w(z)^2) \sin^2[kz - \tan(z/z_0) + k(x^2 + y^2)/2R(z)]$$

where w_0 is the waist, k is the wave number, $w(z) = w_0(1 + z^2/z_0^2)^{1/2}$, $R(z) = z + z_0^2/z$ and $z_0 = kw_0^2/2$. As, under the current experimental conditions, the positions of the individual ions and the phase of the cavity field are not correlated, N can be well approximated by $N \equiv \rho_0 \int_{V_{\text{Coul}}} \psi^2(\mathbf{r}) d\mathbf{r}$, where V_{Coul} is the volume of the Coulomb crystal and ρ_0 is the uniform density of ions in the Coulomb crystal³¹. For the trap used here²⁶ $\rho_0 = (6.03 \pm 0.08) \times 10^3 \times U_{\text{eff}}^2 [\text{V}^2] \text{ cm}^{-3}$, and in the experiments ρ_0 was varied between $1 \times 10^8 \text{ cm}^{-3}$ and $6 \times 10^8 \text{ cm}^{-3}$. The Coulomb crystal volume is determined from the projection fluorescence images and the known rotational symmetry of the crystals. With the typically low-aspect ratio crystals used in the experiments, the total relative uncertainty in N is 2–5%.

Coherence time measurements. Given the magnetic field configuration and the optical pumping preparation, the collective coupling of the crystal with the circularly polarized probe after a time τ is dependent on the population in the $m_J = +3/2$ and $m_J = +1/2$ Zeeman substates of the $3d^2D_{3/2}$ -level. The cooperativity $C(\tau)$ is consequently expected to vary as $a + b\cos(\omega_L\tau) + c\cos(2\omega_L\tau)$, where the Larmor frequency is given by $\omega_L = (\omega_x^2 + \omega_z^2)^{1/2}$, with ω_x and ω_z being the Larmor frequencies induced by the B_x and B_z magnetic field components, respectively, and the parameters a, b, c are functions of B_x, B_z and the optical pumping efficiency. The coherence time is evaluated by fitting the measured data to the above expression with all of the oscillating terms multiplied by a decaying function being either exponential, expected if the decoherence processes give rise to a homogeneous broadening of the energy levels, or Gaussian, obtained for inhomogeneous broadening decoherence processes. Owing to current technical limitations related to the data acquisition at long probe pulse delays, it has not been possible to discriminate between the two types of process.

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