Cooperative Lamb Shift in a Mesoscopic Atomic Array

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According to quantum electrodynamics, the exchange of virtual photons in a system of identical quantum emitters causes a shift of its energy levels. Such shifts, known as cooperative Lamb shifts, have been studied mostly in the near-field regime. However, the resonant electromagnetic interaction persists also at large distances, providing coherent coupling between distant atoms. Here, we report a direct spectroscopic observation of the cooperative Lamb shift of an optical electric-dipole transition in an array of Sr^+ ions suspended in a Paul trap at inter-ion separations much larger than the resonance wavelength. By controlling the precise positions of the ions, we studied the far-field resonant coupling in chains of up to eight ions, extending to a length of 40 μm. This method provides a novel tool for experimental exploration of cooperative emission phenomena in extended mesoscopic atomic arrays.

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In quantum electrodynamics, the interaction of individual atoms with electromagnetic vacuum fluctuations brings about spontaneous emission and shifts the atomic energy levels [1]. As pointed out by Dicke [2], the presence of additional proximate atoms changes the character of the radiative decay dynamics dramatically, giving rise to superradiance phenomena and shifting the energy levels of the compound system [3–5]. Such collective energy level renormalization, known as the cooperative Lamb shift [6–9], can be thought of as resulting from an effective resonance interaction, transferring the excitation between individual emitters via emission and reabsorption of virtual photons. In this context, the superradiance effects and the transition frequency shift originate, respectively, from the imaginary and real parts of the resonant dipole-dipole interaction (RDDI).

RDDI has been observed in diverse systems, ranging from an x-ray magnetic dipole transition in ^{57}Fe nuclei [7] to optical dipole-dipole interaction in a gas of Rb atoms [8], to microwave domain transitions in Rydberg atoms [10–12] and superconducting qubits [13]. In free space, RDDI is predominantly observed as a near-field phenomenon, as the coupling decays cubically with distance when the interatomic separation is much smaller than the transition wavelength [14]. At these small distances, RDDI dominates over relaxation processes, which enabled its observation in pairs of individual quantum emitters such as two fluorescent molecules separated by 12 nm, embedded in a dielectric film [15]. The strong near-field RDDI has also been utilized to prevent the transition of more than one Rydberg atom to the excited state, bringing about the phenomenon known as Rydberg blockade [16–18].

Although RDDI has been mostly studied as a near-field coupling mechanism, it has a long-range term scaling inversely proportional to the distance, which becomes dominant at interatomic separations much greater than the transition wavelength. Detection of RDDI in the far-field regime enables precise measurement of cooperative effects under controlled conditions, in the absence of other types of interactions between the emitters. The far-field coupling regime was first explored by Brewer and DeVoe [19], who detected superradiance in a system of two trapped ions in a Paul trap. Far-field RDDI also plays a central role in a variety of superradiance phenomena, such as the emergence of directionality, which occur in extended mesoscopic samples [20–26].

Here, we experimentally investigate RDDI in a mesoscopic array of atomic ions separated by distances much larger than the transition wavelength. To this end we perform a direct spectroscopic measurement of the cooperative Lamb shift of the $5S_{1/2} \leftrightarrow 5P_{1/2}$ optical dipole transition frequency in a system of several Sr^+ ions suspended in a linear Paul trap. Varying the number of trapped ions from two to eight, we carry out the first observation of cooperative emission phenomena in a mesoscopic array of coupled quantum emitters. The high degree of isolation from the environment and the precise control over the positions of emitters achievable in radio-frequency (rf) ion traps allowed for unobstructed detection of cooperative effects even with an array length that extends to approximately a hundred times the resonant wavelength.

First, we consider cooperative shifts in a system of two trapped ions. A level scheme of the system, taking into account only the ground state $5S_{1/2}$ denoted by $|g\rangle$ and the $5P_{1/2}$ excited state denoted by $|e\rangle$, is shown in Fig. 1(a). Both states have a total angular momentum of $\hbar/2$; thus, each single ion electronic state has two corresponding spin states. The two-ion system therefore has 16 states: four ground states $|gg\rangle$, eight singly excited states $|eg\rangle \pm |ge\rangle$,

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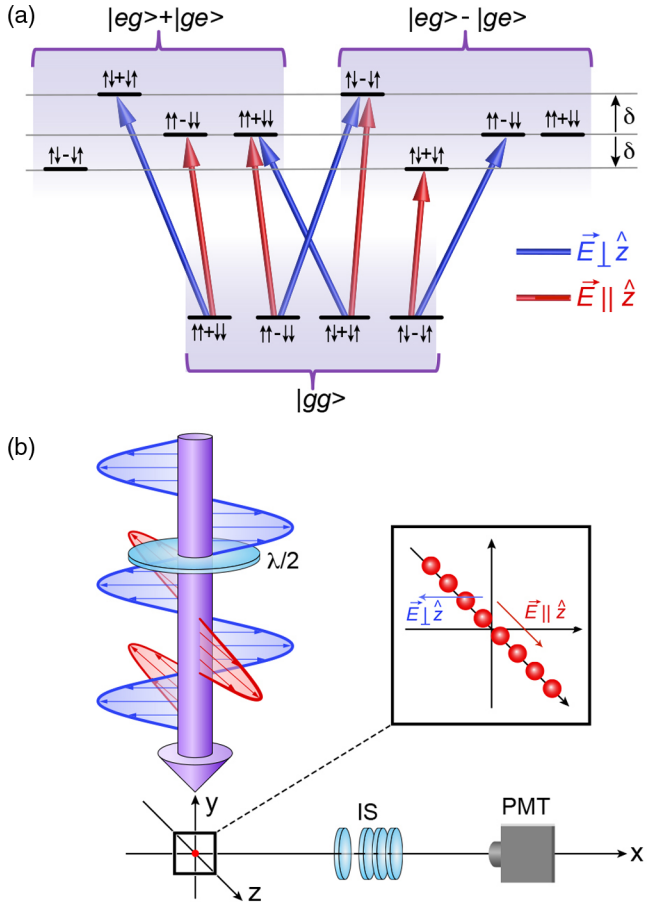

FIG. 1 (color online). (a) Energy level diagram of a two-ion system. $|g\rangle$ and $|e\rangle$ denote electronic ground and excited states. $(\uparrow)$ and $(\downarrow)$ denote the ion spin projection on the trap axis. The states $\frac{1}{2}(|eg\rangle + |ge\rangle)(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ and $\frac{1}{2}(|eg\rangle - |ge\rangle)\times(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$, which turn into themselves upon excitation transfer from one ion to the other $|e\rangle \Leftrightarrow |g\rangle$ and simultaneous spin exchange $\uparrow \Leftrightarrow \downarrow$, are shifted by δ given by Eq. (2). The states which acquire a minus sign upon such exchange, $\frac{1}{2}(|eg\rangle \pm |ge\rangle)(|\uparrow\downarrow\rangle \mp |\downarrow\uparrow\rangle)$, are shifted by $-\delta$. (b) System geometry. Two or more Sr^+ ions are trapped in the center of a linear Paul trap (trap electrodes not shown). The ions form a chain along the trap axis $\hat{z}$. A linearly polarized probe beam, propagating along the $\hat{y}$ direction, illuminates the ions uniformly. The probe beam polarization can be rotated using a $\lambda/2$ plate. In our experiments, the beam was polarized either along the trap axis (red arrows), or orthogonal to it, in the $\hat{x}$ direction (blue arrows). The light scattered by the ions was collected by the imaging system (IS) and detected by a photomultiplier tube (PMT).

and four doubly excited $|ee\rangle$. For the purposes of discussing the energy level arrangement and the cooperative shifts in a two-ion system, we assume that magnetic field is sufficiently small that we can disregard the Zeeman splitting of the levels [27].

In the absence of interaction, all of the eight singly excited states are degenerate. The RDDI creates a coupling between the singly excited states and lifts this degeneracy. The resulting energy splitting is the cooperative line shift. The strength of the RDDI is generally given by the expression [4,14]

$$V_{iqjs} = \frac{k^3}{4\pi\epsilon_0\hbar}\left[-(\mathbf{d_{ij}}\cdot\mathbf{d_{qs}} - (\hat{\mathbf{r}}\cdot\mathbf{d_{ij}})(\hat{\mathbf{r}}\cdot\mathbf{d_{qs}}))\frac{\cos(kr)}{kr} + (\mathbf{d_{ij}}\cdot\mathbf{d_{qs}} - 3(\hat{\mathbf{r}}\cdot\mathbf{d_{ij}})(\hat{\mathbf{r}}\cdot\mathbf{d_{qs}}))\times\left(\frac{\sin(kr)}{(kr)^2} + \frac{\cos(kr)}{(kr)^3}\right)\right], \quad (1)$$

where $\mathbf{d_{ij}}$ and $\mathbf{d_{qs}}$ are the transition dipole moments of the two ions, one transferred from state i to j, and the other from q to s. Here, $k = 2\pi/\lambda$, $\hat{\mathbf{r}}$ is the unit vector in the direction connecting the two ions and r is the distance between them.

The $1/r$ long-range first term of Eq. (1) describes the far-field coupling, whereas the other two correspond to the near-field interactions. At the relatively large inter-ion distances achievable in our ion trap, $r \simeq 5\ \mu\text{m} \simeq 12\lambda$, the far-field coupling dominates.

The spin dependence of this interaction can be intuitively explained in terms of emission and absorption of virtual photons. Choosing the spin quantization along the trap axis, one observes that only $\sigma_\pm$ photons can be emitted from one ion towards the other, while π photons are not emitted in this direction. At the same time, momentum conservation requires that the ion spin must be flipped whenever a $\sigma_\pm$ photon is emitted or absorbed. These considerations lead to the energy level splitting shown in Fig. 1(a). The energy levels of symmetric combinations of the interacting states, $\frac{1}{2}(|eg\rangle + |ge\rangle)(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ and $\frac{1}{2}(|eg\rangle - |ge\rangle)(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$, are shifted by δ, where

$$\delta(r) = -\frac{3}{8}A_{\uparrow\downarrow}\frac{\cos(kr)}{kr} + \mathcal{O}(kr^{-2}). \quad (2)$$

Here, $A_{\uparrow\downarrow} = 2/3A_0$ is the oscillator strength of the spin-flipping transition, the factor $2/3$ is a Clebsch-Gordan coefficient, and $A_0 = 20.05(48)$ MHz is the total oscillator strength of the $5S_{1/2} \leftrightarrow 5P_{1/2}$ transition [28]. The two antisymmetric combinations $\frac{1}{2}(|eg\rangle \pm |ge\rangle)(|\uparrow\downarrow\rangle \mp |\downarrow\uparrow\rangle)$, are shifted by $-\delta$. The four singly excited states with parallel spin projections $\frac{1}{2}(|eg\rangle \pm |ge\rangle)(|\uparrow\uparrow\rangle \pm |\downarrow\downarrow\rangle)$ do not participate in the far-field interaction and are not shifted.

A probe beam polarized orthogonally to the trap axis causes transitions from the four equally populated ground states to a set of excited states shown by the blue lines in Fig. 1(a). The resulting mixture of excited states includes two levels shifted by $\delta(r)$ and two unshifted. The center of the observed spectral line is given by the mean shift of the four transitions, $f(r) = \frac{1}{2}\delta(r)$. If instead the probe beam is polarized along the trap axis, it excites a mixture of states shown by red lines in Fig. 1(a), of which one is shifted by δ, another by $-\delta$, and the rest are unshifted. In this case, the

excited states have zero average shift, and, in contrast to excitation with orthogonal polarization, no line center shift is expected.

The magnitude of the cooperative shift given by Eq. (2) gives a peak-to-peak frequency shift of $\delta \simeq 130$ kHz at a distance of about 5 μm, much smaller than the natural transition line width $\Gamma_0 = 21.5$ MHz [28]. The shifted and unshifted states of the two-ion system are therefore spectrally unresolved. Nevertheless, the cooperative shift can be probed by detecting the shift of the line center, which can be determined with an accuracy much greater than the linewidth, provided that the resonance spectrum can be measured with a high signal-to-noise ratio.

To measure the resonance line shifts, we used an experimental apparatus shown schematically in Fig. 1(b). The ions were suspended in a linear Paul trap, cooled close to the Doppler limit, and prepared in the $S_{1/2}$ ground state. The distance between the ions was varied by tuning the tightness of the axial confinement in the trap. A weak probe beam close to resonance with the $5S_{1/2} \leftrightarrow 5P_{1/2}$ transition was aligned orthogonally to the trap axis, so that the two ions experienced the same phase of the light wave. The probe beam intensity, stabilized by a feedback circuit, was set well below saturation.

The detection sequence consisted of a cooling and repumping procedure interlaced with an 8 μs probe pulse, during which all other light was extinguished and the scattered probe photons were collected by an optical system and detected using a photomultiplier tube. The frequency of the probe beam was scanned across the $5S_{1/2} \leftrightarrow 5P_{1/2}$ transition using an acousto-optical frequency shifter, producing a Lorentzian-shaped line profile with a width of 24.6 MHz. Finding the centroid of the spectral line, we were able to determine the transition frequency with an accuracy of about 50 kHz, limited by the frequency drifts in our apparatus. To overcome this limitation, we resorted to a relative measurement, switching cyclically between different inter-ion distances, spending 30 s at every point. The long-term drifts of the system are thus shifting the measured resonance frequency equally for all the measurement points without affecting the relative frequency shift between different distances [27].

The distance dependence of the $5S_{1/2} \leftrightarrow 5P_{1/2}$ transition frequency for two ions is shown in Fig. 2. With integration time of 8.6 hours per point, the measurement reached an average statistical uncertainty of 2.2 kHz, which is a factor of 10^4 smaller than the transition line width. Since in our scheme only the relative frequency shifts between distances are measured, the average frequency shift of all distance points was set to the theoretically predicted value of $\bar{f}_0 = \frac{1}{2}\langle \delta(r_i) \rangle$, where r_i are the distances at which the measurements were carried out. The root-mean-square deviation of the measured points from the theoretical curve of 2.15 kHz suggests no statistically significant discrepancy

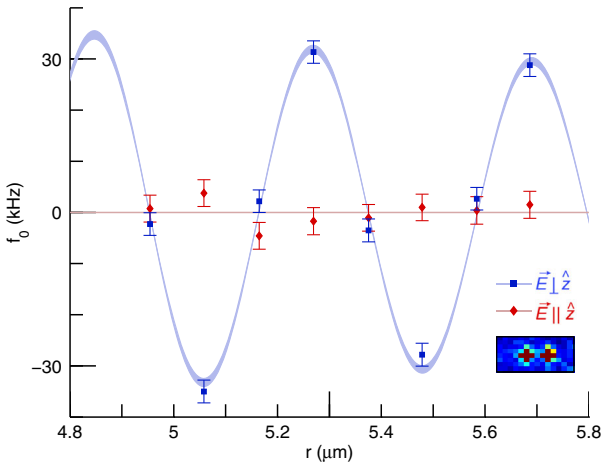

FIG. 2 (color online). Cooperative line shift in a system of two Sr^+ ions. The data points show the measured relative frequency shift as a function of distance between the ions. The blue squares and red diamonds correspond to the probe beam polarization orthogonal and parallel to the trap axis, respectively. The light blue line shows the theoretical shift of $\frac{1}{2}\delta(r)$ with $\delta(r)$ given by Eq. (2). The width of the line reflects the uncertainty in the oscillator strength value. The error bars represent one standard deviation statistical uncertainty. The observed peak to peak spectral shift is approximately 2×10^{-3} of the line width.

with theory. We have also performed the cooperative shift measurement with the probe beam polarized along the trap axis, in which case the cooperative shift is predicted to vanish. Indeed, the measurement results shown as red diamonds in Fig. 2 demonstrate no distance-dependent frequency shift for the parallel polarization of the probe beam.

The good agreement of data with theory suggests we can use this measurement to extract the magnitude of the oscillator strength. Fitting the perpendicular polarization data to Eq. (2) with oscillator strength (A_0) as a single free parameter gives: $A_0^{\mathrm{fit}} = 19.71(88)$ MHz in good agreement with the previously measured value: $A_0 = 20.05(48)$ MHz [28].

The cooperative shift measurement can be extended to a system of several quantum emitters by loading additional ions into the rf trap. The ions arrange themselves in a line with the axial positions given by $r_m = t_m p$, where t_m are constant normalized distances determined by the competition between the harmonic trap potential and the Coulomb repulsion [29]. The scale coefficient p is controlled by the trap stiffness. The inter-ion distances are determined analytically from the axial center-of-mass mode frequency which we measured independently [27].

Similarly to the case of two ions, the role of the Zeeman structure is reducing the observed cooperative shifts by a factor of 1/2. Disregarding the spin structure, the symmetric excited state of M ions created by the weak probe beam can be described as

$$|\psi\rangle = \frac{1}{\sqrt{M}} \sum_{i=1}^{M} |g_1 \ldots e_i \ldots g_M\rangle, \tag{3}$$

and the RDDI Hamiltonian is given by $\hat{V} = \frac{1}{2}\sum_{i\neq j}\delta(r_i - r_j)[\sigma_{+i}\sigma_{-j} + \sigma_{-i}\sigma_{+j}]$, where $\delta(r)$ is given by Eq. (2), $\sigma_{+i} = |e_i\rangle\langle g_i|$ and $\sigma_{-i} = \sigma_{+i}^{\dagger}$. For a chain of $M > 2$ ions, $|\psi\rangle$ is no longer an eigenstate of the interacting system. The observed line shape is given by a weighted sum of Lorentzian resonances, each representing an eigenstate of $\hat{V}$ and shifted by the corresponding eigenenergy. Since the peaks of individual eigenstates are unresolved, the observed line shape is a single Lorentzian-like spectral profile with an apparent shift given by a weighted sum of the eigenstate shifts. This shift can be expressed as the expectation value of the interaction in the excited state [27],

$$\delta_M = \langle\psi|\hat{V}|\psi\rangle = \frac{1}{M}\sum_{i\neq j}\delta(r_i - r_j). \tag{4}$$

The results of the cooperative Lamb shift measurements for three to eight ions are presented in Figs. 3(a)–(f). The data are compared to the theoretically predicted distance dependence curves of Eq. (4) shown by the light blue lines.

The experimental results demonstrate a good agreement with theory. In the three ion case, the chain being equidistant, the contributions of the three ion pairs are in-phase and add constructively at periodic intervals, producing sharp peaks in the distance dependence of the line shift. For an equidistant chain of more than three ions, the periodic peaks will grow sharper with increasing number of ions. The predicted line shift of periodic ion chains is marked by the grey lines in Figs. 3(b)–(f). However, in the harmonic ion trap a chain of more than three ions is no longer equidistant, so the contributions of different ion pairs have incommensurate spatial frequencies. The distance dependence of the shift thus exhibits beating between the spatial frequencies corresponding to all ion pairs. We chose the distance range in our experiments so as to maximize the visibility of the peaks for a given number of ions.

As the features in the distance dependence of the cooperative shifts get sharper, the position uncertainty caused by thermal motion of the ions becomes significant and causes a washing out of the sharp spectral features. The deviation from theory is more pronounced at larger distances, which require smaller axial trap stiffness leading to a greater position uncertainty. Nevertheless, the fact that the data points follow the sharp features and the nontrivial shapes of the theoretical distance dependence of the shifts demonstrates that the entire ion chain participates in the interaction.

The cooperative line shifts investigated here are closely related to the resonance shifts observed in a system

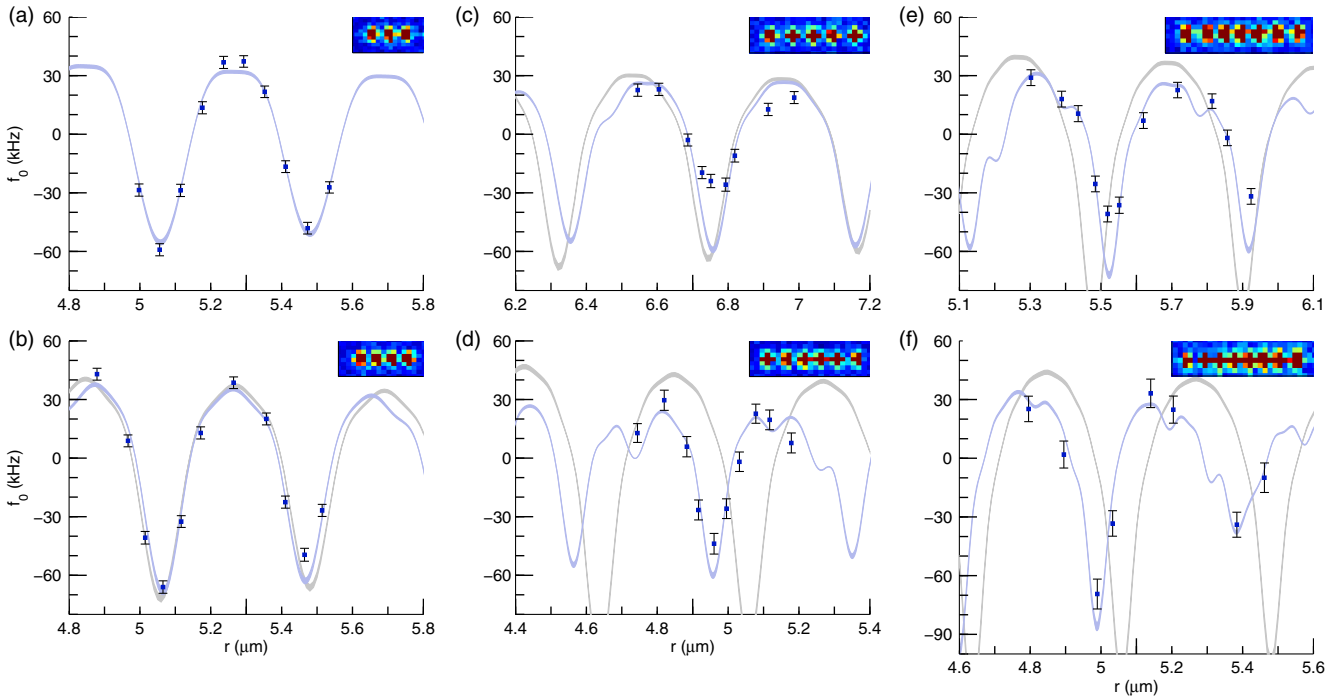

FIG. 3 (color online). Cooperative shift in a linear chain of several ions. The panels (a)–(f) correspond to chains of three to eight ions. In each panel, the cooperative shift is measured as a function of the distance r between two adjacent ions closest to the middle of the chain. The light blue lines show the theoretical shift of Eq. (4) [30]. The error bars represent one standard deviation statistical error. Both the shift scale and the distance scale are the same in all panels. The grey lines, shown for comparison, represent the theoretical shift for equidistant chains. The width of theoretical lines represents the uncertainty in the oscillator strength value.

comprising an ion and a mirror [31]. In such a system, instead of interacting with other ions, the ion interacts with its own mirror image, leading to a shift of the resonance frequency [32,33]. Fundamentally, both types of shifts arise from the same mechanism, i.e., emission and immediate reabsorption of a virtual photon, similarly to the original Lamb shift [1]. The mirror-ion experiments are related to the present work also on another level. Recently, the same group has shown that such ion-mirror systems can form a cavity, with the ion serving as the second cavity mirror [34]. In context of that work, in our experiment the ions can be considered as playing the role of mirrors, reflecting the fields created by other ions.

In summary, we have detected spectroscopically a frequency shift associated with energy transfer between distant atoms. We have demonstrated cooperative effects in which up to eight atomic ions collectively participated. The scope of our work is limited to small emitter arrays and singly excited states. However, the approach demonstrated here opens the door for spectroscopic studies of larger ion crystals that are common in ion trapping experiments, as well as to investigations of quantum physics beyond single-photon excitations. These experiments open the door to the research of far-field cooperative emission phenomena in controllable mesoscopic systems.

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