Spin Correlations as a Probe of Quantum Synchronization in Trapped Ion Phonon-Lasers

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We investigate quantum synchronization theoretically in a system consisting of two cold ions in microtraps. The ions’ motion is damped by a standing-wave laser whilst also being driven by a blue-detuned laser which results in self-oscillation. Working in a non-classical regime, where these oscillations contain only a few phonons and have a sub-Poissonian number variance, we explore how synchronization occurs when the two ions are weakly coupled using a probability distribution for the relative phase. We show that strong correlations arise between the spin and vibrational degrees of freedom within each ion and find that when two ions synchronize their spin degrees of freedom in turn become correlated. This allows one to indirectly infer the presence of synchronization by measuring the ions’ internal state.

Introduction. Two macroscopic self-oscillators synchronize when their relative phase locks to a fixed value [1]. Important studies of synchronization effects were carried out using lasers [2], with arrays of Josephson junctions [3] and over the last few years much attention has been devoted to exploring synchronization in micromechanical oscillators [4]. Recently, theoretical work has begun to explore synchronization in the quantum regime [5–14]: the formation of a relative phase preference between two (or more) weakly coupled quantum oscillators operating in a regime far from the classical correspondence limit. Differences between classical and quantum predictions for the synchronization of van der Pol oscillators have been identified in the case where the oscillators are only weakly excited [5]. Nevertheless, many important questions about quantum synchronization remain open, such as how it should be quantified and how it can best be probed experimentally.

Cold ions in microtraps provide a natural platform for exploring synchronization in the quantum regime [5]. The generation of self-oscillations in the motional state of ions, phonon-lasing, has already been observed [15]. Furthermore, precise control of trapping potentials of the individual ions can now be achieved with microtraps [16] allowing the vibrational frequencies of individual ions and the coupling between different ions to be tuned. Here, we investigate synchronization in two trapped ion phonon-lasers which are pumped in a similar way to that demonstrated in recent experiments [15].

We identify a parameter regime where phonon-lasing of an individual ion occurs with just a few quanta leading to a non-classical state of the phonons and investigate the emergence of synchronization in this regime when a weak inter-ion coupling is introduced (weak as it is the slowest time scale in the system). Our model includes two of the electronic levels of the ions used in the pumping process (which we refer to as ‘spin’), allowing us to uncover strong correlations which arise between the electronic and vibrational degrees of freedom of the individual ions. We study the degree of synchronization as the strength and detuning of the pumping lasers are varied by calculating the probability distribution for the relative phase of the ion’s phonons. Lastly we show that synchronization between the ion’s vibrational degrees of freedom can lead to correlations between the ‘spins’ of the two ions. Indeed, observation of spin-correlations form a sufficient and convenient method of inferring synchronization between two phonon-lasers.

Trapped Ion Setup. A sketch of the system we study is shown in Fig. 1. Each ion is in a microtrap with frequency \( \omega_j=1,2 \), which are set to be resonant with the first blue sideband transition. We adiabatically eliminate the internal electronic states involved with the damping as this happens on a very fast time scale. We assume that the ions are tightly confined and that we are in the Lamb-Dicke regime. This allows us to make a rotating wave approximation and ignore spontaneous-emission-recoil noise (see Supplemental Material). The spin-phonon coupling then simplifies to the anti-Jaynes-Cummings model [19]. The vibrational dynamics of the two ions weakly interact through a linear coupling of

![FIG. 1. (a) Trapped ion setup. Each ion is damped at a rate \( \Gamma \) by a standing-wave laser and driven by a blue-detuned laser of strength \( \Omega_j=1,2 \). The phonons have a dipole interaction of strength \( J \) and the trap frequencies are \( \omega_1 \) and \( \omega_2 \). (b) Internal electronic states of each ion. The ‘spin’ states are pumped by a laser blue-detuned by frequency \( \omega_j=1,2 \) and undergo spontaneous emission at a rate \( \gamma \).](image)
pressure of their spins. The spin-phonon correlations are correlated with phonon numbers, which is confirmed by calculating the correlations, given beneath the steady state of the system. The steady state of the system is given by
\[
\langle n \rangle = \gamma / 2 \Gamma - \gamma^2 / 20^2
\]
where \( n = a^\dagger a \).

Individual Ions. A prerequisite for synchronization is that each individual ion undergoes self-oscillations in their motion, so-called phonon lasing. When the phonons are driven sufficiently strongly to overcome the damping, \( \Omega^2 > \gamma \Gamma \), the mean-field equations of motion show a limit-cycle solution with \( \langle n \rangle = \gamma / 2 \Gamma - \gamma^2 / 20^2 \) where \( n = a^\dagger a \). We have numerically confirmed these mean-field predictions by finding the steady state of an ion for fixed driving \( \Omega \) and decreasing damping rate \( \Gamma (\Delta = 0) \) and \( J = 0 \). We characterize the onset of phonon-lasing with two quantities, the average phonon number \( n_a \) and the phonon number which is most likely to be observed \( n_m \). In Fig. 2(a) we see both these parameters get larger as the damping is decreased. Onset of the lasing is also visible in the Mandel-Q parameter, \( Q = (\langle n^2 \rangle - \langle n \rangle^2) / \langle n \rangle - 1 \). Moreover we see sub-Poissonian statistics around the lasing transition; this is because we are using a single two-level system for the pump. In the following we investigate synchronization in a quantum regime where \( \Omega / \gamma = 1 \) and \( \Gamma / \gamma = 1 / 3 \); here \( n_a = 1.2 \), \( n_m = 1 \) and \( Q = -0.1 \). The steady-state Wigner distribution [21] (for these parameters is shown in Fig. 2(b)). It has a 'doughnut' shape, as the state has a non-zero average amplitude, but no phase preference.

Before investigating synchronization in coupled ions, we examine the correlations that build up between the spin and phonon degrees of freedom in an individual ion due to their strong coupling. We later exploit these correlations to show how the presence of synchronization between the ions’ phonons can be inferred through measurements of their spins. The spin-phonon correlations become apparent in Fig. 2(c), where we plot the Wigner distribution of the density matrix for an individual ion after projection with one of Pauli operator eigenstates. Specifically, we apply \( P^{\alpha, \pm} \), where \( \sigma^\alpha P^{\alpha, \pm} = \pm P^{\alpha, \pm} \) and \( (P^{\alpha, \pm})^2 = P^{\alpha, \pm} \), to the steady state of the system: \( \rho_{ss}^{\pm, \alpha} = \text{Tr}_{s}[P^{\alpha, \pm} \rho_{ss}] \). We see that the projections onto the eigenstates of \( \sigma^x \) are correlated with phonon number, but not phase. Interestingly, we see some negativity in the Wigner distribution after projection with \( P^{\alpha, -} \), which provides further evidence that we are in the quantum regime. On the other hand, projections onto the eigenstates of \( \sigma^x \) are correlated with the phase of the phonons, but not the number. We confirm these observations by studying the correlators \( C(\sigma^x, a) \) and \( C(\sigma^z, n) \) which relate to the spin-phonon phase and number correlations respectively (shown in Fig. 2). The correlation between two operators \( X \) and \( Y \) is defined as \( C(X, Y) = \langle XY \rangle - \langle X \rangle \langle Y \rangle \). As the spin-phonon coupling is strong, we do not refer to this as synchronization, but rather as ‘locking’ [22]. This locking can also be derived from a mean-field treatment (see Supplemental Material).

Coupled Ions. We now consider how synchronization arises for two weakly coupled ions. Classically synchronization originates from the development of stable fixed points in the equation of motion for the relative phase. For a quantum system, our intuition suggests that there exists some relative-phase distribution which is not flat...
when the ions are synchronized. A candidate phase distribution is the Wigner distribution after integrating over the radial and total phase coordinates [5]. But, in general, a distribution based on a quasi-probability distributions is not unique as other representations could be used [24], which would give different results. We circumvent this ambiguity by directly calculating a relative phase distribution from the density matrix using phase states [24]:

\[
P(\phi) = \int_0^{2\pi} d\phi_1 d\phi_2 \delta(\phi_1 - \phi_2 - \phi) \langle \phi_1, \phi_2 | \rho_{ss}^{\phi} | \phi_1, \phi_2 \rangle
\]

\[
= \sum_{n,m=0}^{\infty} \sum_{d=m}^{\infty} \frac{e^{i(m-n)\phi}}{2\pi} \rho_{ss}^{\phi} |d-m, m\rangle
\]

where \( |\phi_j \rangle = \sum_{n=0}^{\infty} e^{i\phi_j n} / \sqrt{2\pi} |n\rangle \) and \( \rho_{ss}^{\phi} = \text{Tr}_s[\rho_{ss}] \) is the steady-state density matrix after tracing over the spins. \( P(\phi) \) is positive and normalized.

We look for a signature of synchronization by calculating the relative phase distribution \( P(\phi) \) from the steady state solution to the master equation when ions are in the lasing regime and weakly coupled: \( J/\gamma = 1/10 \). We plot \( P(\phi) \) in Fig. 3 with different values for \( \Delta \) and \( \Omega_1/\Omega_2 \):

Firstly, we consider the symmetric case (\( \Delta = 0 \) and \( \Omega_1/\Omega_2 = 1 \)) for which \( P(\phi) \) is shown in Fig. 3(a). The ions show signatures of synchronization as \( P(\phi) \) is not flat, in fact the distribution is bimodal and \( \pi \)-periodic. This bimodal feature is analogous to the bistability typically seen in synchronized classical systems with inertial coupling [26].

Next we consider detuning of the oscillators frequencies (see Fig. 3(b)). Here we see the maximum height of \( P(\phi) \) gets smaller as it approaches a flat (unsynchronized) distribution. The \( P(\phi) \) stays exactly \( \pi \)-periodic and bimodal during this process, although the phase of the distribution does shift. Loss of synchronization due to detuning is also seen in classical systems [26]; but it is typically a sharp transition (in the absence of thermal noise), while we see in the quantum case it is smooth [4, 8].

Lastly, when one of the ions is pumped more strongly than the other such that \( \Omega_1 > \Omega_2 \) (while \( \Delta = 0 \)), we find that \( P(\phi) \) changes continuously from being bimodal to unimodal (see Fig. 3(c)). Such transitions between monostable and bistable synchronized states have also been observed in classical systems with unequal driving [27].

\textbf{Measure of Synchronization.} It would be convenient to have a single parameter that quantifies the strength of synchronization for a given \( \rho_{ss} \). We therefore propose a simple measure based on the relative phase distribution:

\[ S = 2\pi \max[P(\phi)] - 1. \]

\( S \) is in essence the peak height of \( P(\phi) \) above a flat distribution. It is a useful measure for synchronization as it is non-zero if and only if \( P(\phi) \) is not flat, which we regard as the signature for the presence of synchronization. We plot \( S \) in Fig. 3(d) and (e) to summarize the strength of synchronization in the system. In Fig. 3(d) we see \( S \) goes to zero as the two oscillators are increasingly detuned and synchronization vanishes.

The measure \( S \) has other useful properties: (i) \( S \) is positive and unbounded, \( S \geq 0 \). Having no maximum value matches what one would expect from alternative synchronization measures such as inverse relative phase variance (1/\( \Delta \phi \)). (ii) \( S \) is maximized by a Dirac delta function. (iii) When only a finite number of Fock states, say \( N \), have non zero amplitude we can put an upper bound on \( S \): \( S \leq N \) (see Supplemental Material). Physically, this implies the strength of synchronization has the potential to be much larger with semi-classical states (large \( N \)) than quantum states (small \( N \)).

\textbf{Synchronization and Spin Correlations.} We have shown that synchronization is present in the motional state of our ions using the distribution \( P(\phi) \) and the related measure \( S \). Sophisticated experimental techniques have been developed to perform full state-tomography of an ion’s phonons which would give access to \( \rho_{ss}^{\phi} \). From which \( P(\phi) \) and \( S \) could be calculated. Nevertheless, the spin-phonon locking seen in Fig. 2(c) suggests we may be able to infer the presence of synchronization indirectly through measurements [19] of the spin degrees of freedom alone.

The spin-phonon locking occurs faster than the phonon-phonon synchronization as the inter-ion coupling provides the longest time-scale in the system. Thus it is reasonable to make a measurement of each ion’s spin and use our \textit{a priori} knowledge of the spin-phonon locking to infer the phase of the oscillators. We focus on the spin-
spin correlation in particular, which we split into two types: ‘number correlations’ (e.g. $C(\sigma_x^1, \sigma_x^2)$ and ‘phase correlations’ (e.g. $C(\sigma_y^1, \sigma_y^2)$ and $C(\sigma_z^1, \sigma_z^2)$).

The phase correlations can be related semi-classically to statistical moments of $P(\phi)$, which in turn give us information about the ion’s state of synchronization. Solving the mean field dynamics for $\langle \sigma_j \rangle$ in terms of $\langle a_j \rangle$ gives $\langle \sigma_j \rangle \sim -ie^{-i\phi}t$ with $\phi_j = \arg\langle a_j \rangle$, in agreement with Fig. 2(c). Although, strictly speaking all correlations are zero in a mean-field calculation, we can use this mean-field equality as an ansatz to describe the mean field dynamics for $\Phi_c, \Phi_x, \Phi_y, \Phi_z$ hence $\Phi_c, \Phi_x, \Phi_y, \Phi_z$ are all zero. This is an example where $\Phi_c, \Phi_x, \Phi_y, \Phi_z$ match $\Phi_c, \Phi_x, \Phi_y, \Phi_z$ respectively. However, these quantities are all zero.

We investigate the connection between spin correlations and synchronization in Fig. 4(a), where we plot $C(\sigma_x^1, \sigma_x^2), C(\sigma_y^1, \sigma_y^2), C(\sigma_z^1, \sigma_z^2)$, $\Phi_c$ and $\Phi_s$, as a function of the detuning $\Delta$ for equal driving strength ($\Omega_1/\Omega_2 = 1$). We plot $S$ for the same parameters in Fig. 4(b) to measure the synchronization strength. The number correlation $C(\sigma_x^1, \sigma_x^2)$ is initially negative and approaches zero as the detuning is increased. This indicates that the phonon-numbers of the oscillators are correlated at small $\Delta$, but does not directly indicate a phase relationship. Note that $C(\sigma_x^1, \sigma_x^2)$ and $C(\sigma_y^1, \sigma_y^2)$ match $\Phi_c$ and $\Phi_s$, respectively. However, these quantities are all zero. This is an example where $\Phi_c, \Phi_s = 0$ which means we do not have sufficient information to conclude whether the ions are synchronized or not. Indeed, $S$ is non-zero in this case. This occurs because $P(\phi)$ is $\pi$-periodic for equal driving, even as the lasers are detuned (see Fig. 3(b)); any phase distribution that is $\pi$-periodic will give $\Phi_c, \Phi_s = 0$ and hence $C(\sigma_x^1, \sigma_x^2) = C(\sigma_y^1, \sigma_y^2) = 0$. Normally one would consider a different statistical moment of the probability distribution to circumvent this issue e.g. if one had two random variables that were uncorrelated in their averages $C(X, Y) = 0$ one could determine $C(X^2, Y^2)$ and possibly find correlations in their variances. Unfortunately the Pauli operator algebra makes such an approach impossible e.g., $(\sigma_x^1)^2 = 1$.

This problem can be overcome by introducing a slight asymmetry in the driving strengths, which breaks the $\pi$-periodic nature of $P(\phi)$ (see Fig. 3(c)). In Fig. 4(c) we plot the same correlations and moments as Fig. 4(a) against detuning, but this time with unequal driving $\Omega_1/\Omega_2 = 5/4$. The spin correlations are now all present, with the phase correlations being much stronger than the number correlations. Even though we are in a quantum regime, we see $C(\sigma_x^1, \sigma_x^2)$ and $C(\sigma_z^1, \sigma_z^2)$ have behavior that follows that of the semi-classical estimates $\Phi_c$ and $\Phi_s$, respectively. As $C(\sigma_x^1, \sigma_x^2)$ and $C(\sigma_z^1, \sigma_z^2)$ are non-zero, we can infer that $\Phi_c$ and/or $\Phi_s$ of the phase distribution are non-zero, which implies the ions are synchronized. This prediction is confirmed by $S$, plotted with the same parameters as (a) and (c) respectively. Here $\Omega_1/\gamma = 1, \Gamma/\gamma = 1/3$ and $J/\gamma = 1/10$.

Detecting correlations on the order of $10^{-3}$ is challenging, but possible with current technology. Projective measurements of the ions’ internal states [19, 28] have been used to estimate observables with a precision over an order of magnitude higher than our requirement [29].

Conclusions and Outlook. We have shown that two phonon-lasing ions undergo synchronization when they are weakly coupled, leading to a bimodal relative phase distribution. Strong correlations develop between the internal, spin, degrees of freedom and the phonons in each ion and when the ions are coupled their synchronization leads to characteristic correlations between the spins. These correlations carry information about the relative phase distribution of the ions and could be used infer the presence of synchronization.

The coupled ion phonon-laser system we consider is a promising model for future studies into quantum synchronization. Correlations between the spins of two ions at different times could be used to probe the dynamics of the synchronization process. Furthermore, the question of whether the non-classical (number-squeezed) phonon states that occur in this system might affect the development of synchronization, leading to significant differences compared to a corresponding semi-classical description [5], remains to be explored.

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MATERIAL EQUATION DERIVATION

Two key approximations are made in the main text when deriving the master equation [Eq. (1)]: the rotating wave approximation (RWA) and a Lamb-Dicke approximation (LDA). Here we verify that these approximations are valid for our choice of parameters.

The dynamics of the internal (electronic) states driven by the standing wave laser is assumed to occur on a much faster time scale than any of the other processes involved and they are adiabatically eliminated (see e.g. [58]) leading to damping of the phonons at a rate $\Gamma$. The master equation for our system before any approximations with regard to the blue-detuned laser driving have been made is \[ \dot{\rho} = L_\epsilon(\rho), \]
with
\[
L_\epsilon(\rho) = -i[H_\epsilon, \rho] + \sum_{j=1,2} \left\{ \frac{\gamma}{2} \int_{-1}^{1} dz W(z) D[e^{iz\sigma_j}]\rho + \Gamma D[a_j] \right\} \tag{4}
\]

$H_\epsilon \equiv \sum_{j=1,2} \left\{ \omega_j (a_j^\dagger a_j - \frac{\sigma_j^z}{2}) - i\Omega (e^{i\varphi_j\sigma_j^z} - e^{-i\varphi_j\sigma_j^z})/2 \right\} + J q_j q_j^\dagger W(z) = (3/4)(1 - z^2), q_j = a_j^\dagger + a_j$ and $D[L](\rho) = L\rho L^\dagger - (L^\dagger L \rho + \rho L^\dagger L)/2$. Here $\Omega$ is the Lamb-Dicke parameter, $\Omega$ is the unscaled driving strength of the laser (related to $\Omega$ in the main text by $\Omega = \eta \Omega$), $W(z)$ is probability distribution for the direction a photon is spontaneously emitted, and other quantities are as defined in the main text.

We first apply the LDA. Assuming $\eta \ll 1$ we expand the exponentials in Eq. (4) retaining only terms up to and including first order in $\eta$. Thus $L_\epsilon(\rho)$ takes the simplified form:
\[
L_\eta(\rho) = -i[H_\eta, \rho] + \sum_{j=1,2} \left\{ \gamma D[\sigma_j^z] \rho + \Gamma D[a_j] \right\}, \tag{5}
\]
with $H_\eta \equiv \sum_{j=1,2} \left\{ \omega_j (a_j^\dagger a_j - \frac{\sigma_j^z}{2}) - \Omega (\sigma_j^\dagger + \sigma_j^z q_j q_j^\dagger)/2 \right\} + J q_j q_j^\dagger$. Next we assume the trapping potentials are tight, so that $\omega_j \gg \Omega, \gamma, \Gamma, \Delta$, then carry out a RWA. A unitary transformation to a rotating frame is performed with $U(t) = \exp(i\omega t \sum_{j=1,2} [a_j^\dagger - \sigma_j^z/2])$ where $\omega = (\omega_1 + \omega_2)/2$ is the compromise frequency. Terms in the transformed master equation rotating at a frequency $\omega$ or higher are then neglected, reducing $L_\eta(\rho)$ from Eq. (5) to
\[
L_\epsilon(\rho) = -i[H_\epsilon, \rho] + \sum_{j=1,2} \left\{ \gamma D[\sigma_j^z] \rho + \Gamma D[a_j] \right\} \tag{6}
\]
with $H_\epsilon \equiv \sum_{j=1,2} \left\{ (-1)^j \Delta (2a_j^\dagger a_j - \sigma_j^z)/4 + \Omega (\sigma_j^\dagger a_j + \sigma_j a_j^\dagger)/2 \right\} + J (a_j^\dagger q_2 + q_2^\dagger a_j)$. $L_\epsilon(\rho)$ is the generator for the master equation presented in the main text.

The validity of our approximations can be checked numerically by calculating the steady state solutions $\rho_{ss}$ generated by the superoperators defined in (4), (5), and (6). Finding the steady state numerically for coupled ions without the rotating wave approximation is demanding as there is a large difference in time scales between the coupling strength $J$ and trap frequencies $\omega_j$. Fortunately, whether the coupling or detuning terms are zero or not does not affect the validity of our approximations, as they are both typically very small compared to $\omega$. Thus we set $J = 0$ and $\Delta = 0$, then check the approximations using a single ion.

Quantities derived from the steady states $\rho_{ss}^\beta$, $\rho_{ss}^r$, and $\rho_{ss}^\alpha$ calculated using superoperators $L_\epsilon$, $L_\eta$, and $L_\epsilon$, respectively, are plotted in Fig. 5. We choose parameter values which match those used for the majority of the main text, in particular, $\Omega/\gamma = 1$ and $\Gamma/\gamma = 1/3$. We set the Lamb-Dicke parameter to $\eta = 0.1$, which corresponds to $\omega_j \approx 2\pi \times 1$ MHz for $^{40}$Ca$^+$. The average phonon number of these steady state solutions is plotted in Fig. 5(a), i.e. $n_{ss}^\beta \equiv \text{Tr}[a_j^\dagger a_j^\dagger \rho_{ss}^\beta]$, with $\beta = \epsilon, \eta, r$. For these parameters the LDA matches the full solution well whilst the RWA typically overestimates the size of limit cycles for relatively low values of $\omega$, but when $\omega$ is sufficiently large, $\omega/\gamma > 400$, we also get very good agreement. To characterize how similar the states are more fully we plot the Fidelity of $\rho_{ss}^\beta$ and $\rho_{ss}^r$ compared to $\rho_{ss}^\alpha$ in Fig. 5(b), i.e.
\[
F(\rho_{ss}^\beta, \rho_{ss}^\alpha) = \text{Tr}[\sqrt{\sqrt{\rho_{ss}^\beta} \rho_{ss}^\alpha \sqrt{\rho_{ss}^\beta}}] \tag{7}
\]
with $\beta = \eta, r$. Again, we see both approximations are in good agreement with the exact master equation for $\omega/\gamma > 400$.

UPPER BOUND ON S

We now outline how an upper bound for the synchronization measure arises when a finite number of Fock
states (specifically $N$) have non-zero occupation. We can place an upper bound on the maximum value of $S$ that $\rho$ can have as follows. The definition of $S$ is:

$$S = 2\pi \max[P_\rho(\phi)] - 1,$$

with

$$P_\rho(\phi) = \frac{1}{2\pi} \sum_{n,m,l}^{N+l} \rho_{m,n} e^{i(d-n)\phi} \rho_{n,n} e^{i(d-m)\phi}.$$  \hspace{1cm} (9)

where $l$ is some arbitrary offset of the state from the vacuum. Our task is to maximize $S$ over all possible density matrices $\rho$. $P_\rho(\phi)$ can always be rotated by changing the phase of the off-diagonal elements of $\rho$ such that the maximum is positioned at $\phi = 0$, i.e. $P_\rho'(0) = \max[P_\rho(\phi)]$. Thus if we want to find the maximum possible value of $S$ it is sufficient to determine the maximum value $P_\rho(0)$ can take. For $\phi = 0$, Eq. (9) reduces to

$$P_\rho(0) = \frac{1}{2\pi} \sum_{n,m,l}^{N+l} \rho_{m,n},$$

where $\rho_{m,n} = \sum_{d=\max(n,m)}^{\min(n+N,m+N)} \rho_{d-n,n} \rho_{d-m,m}$ is a $(N+1) \times (N+1)$ positive semi-definite matrix with unit trace, $\sum_{n=0}^{N} \rho_{n,n} = 1$. $P_\rho(0)$ is therefore the sum over all elements of a positive semi-definite matrix. However, the off-diagonal elements of a positive semi-definite matrix cannot be larger than the diagonal elements, $|\rho_{m,n}| \leq \sqrt{\rho_{m,m} \rho_{n,n}}$. This bound means that $P_\rho(0)$ is maximized when all the diagonal elements have an equal value, as this allows the off-diagonal elements to contribute maximally. As the trace must be unity, the values for $\rho_{m,n}$ that maximize $P_\rho(0)$ are $\rho_{m,n} = 1/(N+1)$. Using these values in Eq. (8) leads to the bound discussed in the main text $S \leq N$.

**SPIN CORRELATIONS**

Finally, we give an approximate semi-classical argument to show how the relationships between the spin correlations and relative phase distribution $P(\phi)$, $C(\sigma_1^z, \sigma_2^z) \propto \Phi_\epsilon \equiv \int d\phi \cos \phi P(\phi)$ and $C(\sigma_1^y, \sigma_2^y) \propto \Phi_\epsilon \equiv \int d\phi \sin \phi P(\phi)$, can be understood. We start by making a semi-classical approximation (e.g. $\langle \sigma_j^+ \alpha_j \rangle \approx \langle \sigma_j^+ \rangle \langle \alpha_j \rangle$), to obtain simple (mean-field) equations for the expectation values of the spin operators of an individual ion (taking $\Delta = 0$ for simplicity),

$$\dot{s}_j = -\gamma s_j + \frac{\Omega}{2} s_j^* \alpha_j$$

$$\dot{s}_j^* = i\Omega(s_j \alpha_j - s_j^* \alpha_j^*) - \gamma(s_j^* + 1),$$

where $\alpha_j = \langle \alpha_j \rangle$, $s_j^- = \langle \sigma_j^- \rangle$ and $s_j^y = \langle \sigma_j^y \rangle$. In the steady state, $s_j = s_j^* = 0$ and we can then solve for $s_j$ in terms of $\alpha_j$: $s_j = (-i\gamma \Omega \alpha_j^*)/(\gamma^2 + 2\Omega^2 |\alpha_j|^2)$ (this is the origin of spin-locking). Writing $\alpha_j$ in terms of phase and amplitude $\alpha_j \equiv r_j e^{i\phi_j}$ gives $s_j \propto -ie^{-i\phi_j}$. Similar steady-state relations follow for $s_j^x = \langle \sigma_j^x \rangle$, $s_j^y \propto \sin \phi_j$, $s_j^z \propto \cos \phi_j$, $s_j^x \propto \sin \phi_j - \phi_j$ and $s_j^y \propto \sin \phi_j + \phi_j$.

When we have two ions, the corresponding products of expectation values of Pauli operators for the two spins lead to different sinusoidal functions of the total and relative phase,

$$s_j^z s_j^x \propto \cos(\phi_1 - \phi_2) - \cos(\phi_1 + \phi_2),$$

$$s_j^z s_j^y \propto -\sin(\phi_1 - \phi_2) - \sin(\phi_1 + \phi_2).$$

In the main text we look at correlation functions $C(X,Y) = \langle XY \rangle - \langle X \rangle \langle Y \rangle$. Strictly speaking, if the semi-classical approximation is correct we would find $C(X,Y) = 0$ for any correlation by definition. However, we can obtain some insight into how the spin correlation functions are related to the underlying phase distributions if we make the ansatz that the semi-classical proportionality between spin expectation values and phonon phase also hold (at least approximately) for the corresponding operators, i.e. we assume $\sigma_j^z \propto -ie^{-i\phi_j}$. We then calculate the expectation values by taking an average over the corresponding steady-state phase distributions (remembering that there is no preferred total phase so its distribution is always flat). This then leads us to the approximate relationships between the expectation values and moments of the relative phase distribution:

$$\langle \sigma_j^z \rangle \propto 0, \langle \sigma_1^y \sigma_2^y \rangle \propto \int d\phi \cos \phi P(\phi), \int d\phi \sin \phi P(\phi).$$

Replacing these into our definition for the correlation functions, we obtain the relations discussed in the main text: $C(\sigma_1^z, \sigma_2^z) \propto \Phi_\epsilon \equiv \int d\phi \cos \phi P(\phi)$ and $C(\sigma_1^y, \sigma_2^y) \propto \Phi_\epsilon \equiv \int d\phi \sin \phi P(\phi)$.

Given the approximations that go into their derivation, we do not expect detailed agreement between the moments $\Phi_\epsilon$, $\Phi_\epsilon$ and the correlation functions $C(\sigma_1^z, \sigma_2^z)$; indeed the agreement seen in Fig. 4 of the main text is only qualitative. Apart from the underlying semi-classical formulation, our derivation also neglected the fact that the constants of proportionality and the precise details of the phase relationship will change as the detuning, $\Delta$, is varied. Nevertheless, the fact that the quantum spin correlations show similar behavior to the moments $\Phi_\epsilon$, $\Phi_\epsilon$ shows us (at least to a first approximation) what information they contain about the relative phase distribution of the system and the argument we have developed above helps to explain why this should be so.