Cavity quantum electrodynamics of strongly correlated dipolar matter

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The structure of solids and their phases is mainly determined by static Coulomb forces while the coupling of charges to the dynamical, i.e., quantized degrees of freedom of the electromagnetic field plays only a secondary role. Recently, it has been speculated that this general rule can be overcome in the context of cavity quantum electrodynamics (QED), where the coupling of dipoles to a single field mode can be dramatically enhanced. Here we present a first exact analysis of the ground states of a dipolar cavity QED system in the non-perturbative coupling regime, where electrostatic and dynamical interactions play an equally important role. Specifically, we show how strong and long-range vacuum fluctuations modify the states of dipolar matter and induce novel phases with unusual properties. Beyond a purely fundamental interest, these general mechanisms can be important for potential applications, ranging from cavity-assisted chemistry to quantum technologies based on ultrastrongly coupled circuit QED systems.

INTRODUCTION

In QED, the relevant dimensionless coupling parameter is the fine-structure constant \( \alpha_{fs} = E_C/E_{ph} \). It can be expressed as the ratio between the Coulomb energy, \( E_C = e^2/(4\pi\varepsilon_0 d) \), of two electrons at a distance \( d \) and the energy \( E_{ph} = \hbar c/d \), which is needed to create a photon confined approximately to the same region in space. The small value of \( \alpha_{fs} \lesssim 1/137 \) already suggests that the quantized modes of the electromagnetic field play a minor role in the physics of atoms, molecules and solids, as confirmed by more rigorous calculations. However, this argument does not necessarily hold in structured electromagnetic environments, such as nanoplasmonic systems or LC circuits, where the energy of a photon can be tuned independently of its wavelength. In this case the coupling between an electric dipole and a quantized field mode is characterized by an effective parameter \( \alpha = \alpha_{fs}(Z/Z_0) \) [1, 2], which can be considerably enhanced by increasing the impedance of the mode, \( Z \), compared to the value in free space, \( Z_0 \). This raises an important fundamental question: Can the properties of matter be influenced by such an artificially boosted coupling to the quantized field, and, if so, how would the properties change?

In view of a growing number of experimental setups where \( \alpha \sim O(1) \) can potentially be reached [3–6], this question has lately gained additional relevance and first observations of cavity-induced modifications of chemical reactions [7], phase transition points [8, 9] and electri-transport [10] have been reported. Moreover, values of \( \alpha \gtrsim 1 \) [11–13] are already accessible in circuit QED, where artificial atoms in form of superconducting two-level systems are coupled to microwave resonators [14, 15]. Already now, such systems offer many intriguing possibilities for investigating basic principles of light-matter interactions in unprecedented coupling regimes. However, due to the complexity of the problem, our understanding of strong-coupling induced modifications of real and artificial matter is still rather poor, even on a conceptual level. While detailed numerical simulations have been performed for simple molecules coupled to a single cavity mode [16–18] or small superconducting circuits [19–22], the analysis of larger systems is usually constrained to idealized collective-spin models [2, 23–27] or to moderate coupling regimes (\( \alpha < 1 \)) [28–32], where no significant modifications of ground and thermal states are expected yet [16, 33–35]. Consequently, still little is known about few- and many-body effects that arise from the direct competition between short-range electrostatic interactions and a non-perturbative coupling to an extended dynamical mode.

In the following analysis we address this open theoretical problem in many-body cavity QED by considering the conceptually simplest scenario of a lattice of interacting two-level dipoles coupled to a single cavity mode. For this system, we use exact numerical calculations to evaluate the ground state properties of a strongly correlated cavity QED system and to identify the key mechanisms that lead to the formation of novel, cavity-induced phases of dipolar matter. By that, this basic study already reveals that there is still a wealth of unexplored phenomena in cavity and circuit QED, which may soon become accessible with further experimental advances in these fields.

CAVITY QED OF INTERACTING DIPOLES

We consider a prototypical cavity-QED system as depicted in Fig. 1(a), where \( N \) anharmonic dipoles are coupled to a single quantized mode of an LC resonator [26]. We approximate the dipoles by two-level systems located at fixed lattice positions \( r_i \) (in units of the lattice spacing \( a \)). The dipoles couple to the electric field \( \vec{E} \) of the cavity with a transition dipole moment \( \vec{\mu} \) and among each
other via static dipole-dipole interactions. Under these assumptions the dynamics of the system is described by the Hamiltonian [2] ($\hbar = 1$)

$$
H_{\text{cQED}} = \omega_d a^\dagger a + \omega_a S_z + g (a^\dagger + a) S_x + \frac{g^2}{\omega_d} S_x^2 + \sum_{i<j} \frac{J_{ij0}^0}{|\vec{r}_i - \vec{r}_j|^3} \sigma_x^i \sigma_x^j + g N \mu_0 \int d^3 \vec{r} \langle \phi_0 | J_0 \cdot \vec{d}(\vec{r}) | \phi_0 \rangle,
$$

where $\sigma_x^i$ denote the Pauli operators at site $i$, $S_x = \sum_i \sigma_x^i/2$ are collective spin operators, and $a$ is the annihilation operator for the field mode.

The cavity affects the dynamics of the dipoles in two different ways. First, in Eq. (1) the dipole frequency, $\omega_d$, and the dipole-dipole couplings, $J_{ij0}$, already include screening effects from the metallic boundaries and can differ considerably from their bare values $\omega_d^0$ and $J_{ij0}^0$ in free space. This behaviour is illustrated in Fig. 1(b-d), which shows that the usual dipole-dipole interactions, $J_{ij0}^0 = J^0/|\vec{r}_i - \vec{r}_j|^3$, become short-ranged and suppressed as the distance $D$ between the plates is decreased. This boundary effect can strongly modify the properties of para- and ferroelectric systems [2, 36–38], but it is of electrostatic origin and not the main focus of this study. Therefore, we simply treat $\omega_d$ and $J_{ij0}$ as arbitrary model parameters and investigate the additional modifications induced by the coupling to the dynamical field mode with frequency $\omega_c \sim \omega_d$. For a sufficiently homogeneous mode, these effects are described by the collective dipole-field coupling $g (a^\dagger + a) S_z$, with a single-dipole coupling strength $g$, and the associated depolarization shift $\sim S_z^2$. This last term ensures that for $\omega_c \to 0$ we recover the correct electrostatic limit, $H_{\text{cQED}} \approx \omega_d a^\dagger a + \sum_{i<j} \frac{J_{ij0}^0}{|\vec{r}_i - \vec{r}_j|^3} \sigma_x^i \sigma_x^j$ [see Methods]. Therefore, although being based on several simplifying assumptions, this model allows us to treat electrostatic and electrodynamical interactions in a fully consistent manner.

**THE GROUND STATES OF CAVITY QED**

The physics of $H_{\text{cQED}}$ and variants thereof has been studied extensively in quantum optics and solid-state physics, but primarily in the regime $g/\omega_c \ll 1$. In this limit, the system may still feature huge collective Rabi splittings of $\Omega_R = gN/\sqrt{N} \sim \omega_c$ in the excitation spectrum [3, 5, 6], but qualitative changes in the ground and equilibrium states are still only perturbative [2, 16, 33–35]. In turn, preliminary studies of $H_{\text{cQED}}$ in the non-perturbative coupling regime, $g/\omega_c \gg 1$, have been restricted to very few spins or the special case of all-to-all interactions $J_{ij} = J$ [2, 21, 35]. This strongly reduces the computational complexity of the problem, but also ignores all non-collective correlations, the influence of the lattice geometry and other essential effects. Here we perform exact, large-scale numerical simulations of finite-sized dipole systems to obtain the ground states of $H_{\text{cQED}}$ without any further approximations [see Methods].

In Fig. 2(a) we first show the ground state phase diagram for $N = 26$ dipoles on a square lattice with nearest-neighbor only couplings $J_{ij} = J \delta_{i,j}$ and $\omega_d = \omega_c$. For $g = 0$ the cavity is completely decoupled and Eq. (1) reduces to the familiar transverse field Ising (TFI) model. In this limit we observe the expected transition from a paraelectric to a ferroelectric or a Néel-ordered phase when $|J|$ exceeds a critical value of $|J^*| \approx 0.7 \omega_d$, which agrees within a few percent with the transition point of the infinite system [39]. Although for finite-size systems symmetries cannot be broken spontaneously and the order parameters are strictly zero, the two ordered phases can be uniquely identified through the correlations between the spins. Thus, to identify the ordered phases, we introduce $\langle \sigma_x \sigma_x \rangle_{\text{ferro}}$ and $\langle \sigma_x \sigma_x \rangle_{\text{Néel}}$ [see Methods], which are nonzero in the corresponding ordered phases, but vanishingly small elsewhere [see right panel in Fig. 2(a)]. For finite $g/\omega_c \lesssim 1$ this picture does not change considerably, except that in the ferroelectric state now also the photon number acquires a large expectation value, $\langle a^\dagger a \rangle \simeq (gN/(2\omega_c))^2$. In the quantum optics literature one commonly refers to such a phase as ‘superradiant’ [23, 24], a notation that we adopt in the following.
FIG. 2. Cavity QED ground states on the square lattice. (a) Phase diagram of $H_{cQED}$ (left panel) and order parameter correlations for a cut at $g/\omega_c = 4$ (right panel). (Left panel) The insets show sketches of the spin configurations in the corresponding phases. The symbols track the phase boundaries estimated for different system sizes $N$, solid (dotted) lines indicate phase transitions (crossovers). (b) Ground state photon number (left panel) and order parameter correlations (right panel) for the transitions from the paraelectric to the subradiant phases with $\langle a^\dagger a \rangle \simeq 0$ along the horizontal dashed lines in (a). For small dipole-dipole interactions, $J/\omega_d$, the collective subradiant regime is stabilized by increasing $g/\omega_c$, while the spins remain disordered (brown curves). For non-zero antiferroelectric interactions, $J/\omega_d > 0$, the system undergoes a phase transition to the Néel subradiant phase, where $\langle a^\dagger a \rangle$ vanishes simultaneously with the onset of antiferroelectric Néel order, $\langle \sigma_x \sigma_x \rangle^{stag} \rightarrow 1$ (pink curves).

Similarly, we refer to the anti-aligned Néel phase with a staggered arrangement of dipoles as ‘subradiant’, since the photon number $\langle a^\dagger a \rangle \simeq 0$ in this state is much smaller than in the fluctuating paraelectric phase.

With increasing $g$ we observe, first of all, a significant cavity-induced reduction of the critical coupling strength $J^* (g)/\omega_d$. More importantly, for $g/\omega_c \gtrsim 3$ the paraelectric phase gradually evolves into a new ‘collective subradiant’ phase with unusual properties [see also Supplementary Section IV]. This phase exhibits no order and $\langle \sigma_x \sigma_x \rangle^{ferro} \simeq \langle \sigma_x \sigma_x \rangle^{stag} \simeq 0$ [see right panel in Fig. 2(a)]. At the same time, also the photon number $\langle a^\dagger a \rangle$ vanishes, indicating that all the dipoles are still anti-aligned. These seemingly contradicting properties can be understood by looking at the limit $J = 0$ and $g \rightarrow \infty$. In this case it can be shown that the ground state of $H_{cQED}$ is the fully symmetric, perfectly anti-aligned state $\langle \psi_{stag} \rangle$, which obeys $S_x (\psi_{stag}) = 0$ and has maximal total spin of $S = N/2$ [21].

FIG. 3. Order parameter fluctuations. Fluctuations for the phase transitions between the disordered and (a) ferroelectric, (b) Néel phases as a function of the dipole-dipole interaction $J/\omega_d$. The peaks are used to estimate the finite-size phase boundaries $J^* (g/\omega_c, N)$ [see also Fig. 2(a)]. The dashed vertical lines indicate the critical point $J^* (g = 0)$ in the thermodynamic limit $N \rightarrow \infty$ [39]. With increasing coupling $g/\omega_c$, we observe a cavity-induced reduction of $|J^*|^/\omega_d$ and a narrowing of the critical region, which is estimated by the width of the peaks.

Compared to a Néel-ordered configuration, this highly entangled state represents an equal superposition of all possible combinations with half of the dipoles pointing along $x$ and the other half into the opposite direction, without any spatial order. Surprisingly, this peculiar collective phase survives to a high degree in the presence of competing short-range interactions and is separated from both ordered phases by a sharp transition.

Although both the collective and Néel ordered phases, are subradiant, a crucial difference between them is visualized in Fig. 2(b). These two plots compare the photon number $\langle a^\dagger a \rangle$ and $\langle \sigma_x \sigma_x \rangle^{stag}$ for fixed $J/\omega_d$, but increasing $g/\omega_c$. For a value of $J = 0.5/\omega_d$ the system then directly transitions from the paraelectric into the Néel phase. This is signified by an increase of $\langle \sigma_x \sigma_x \rangle^{stag}$ and simultaneously, or better to say as a consequence of that, also the photon number vanishes. For $J = 0$ the staggered polarization is always vanishingly small, but the dipoles still completely decouple from the photons for large $g/\omega_c$. The formation of such a collective subradiant phase is thus much more subtle than an interaction induced spatially ordered anti-alignment of dipoles.

While the numerically accessible dipole numbers, $N$, are not large enough to evaluate the critical scaling behaviour along the phase boundaries $J^* (g)$ between the ordered and disordered phases, we can compare the critical features with the results at $g = 0$, where it is well-known that the phase transitions are continuous and in the $(2+1)$D Ising universality class. In Fig. 3 we show that for all $g/\omega_c$, the different phases are clearly delimited by sharp peaks in the fluctuations of the order parameters [see Methods]. With increasing $g/\omega_c$, the width of these peaks shrinks, which indicates a narrowing of the critical region. Moreover, for the transition between
ORDER AND FLUCTUATIONS

From the analysis above we can extract two basic cavity-induced many-body effects, namely the stabilization of phases with pre-existing order and the suppression of fluctuations in the disordered phase through the formation of highly entangled collective states. One thus expects that also in general cavity-induced modifications will be most significant in situations, where strong fluctuations occur already in the bare system. As a prototypical example we now consider the ground state phases of $H_{cQED}$ for repulsive dipoles on a triangular lattice, where we assume nearest-neighbor interactions $J_{ij} = J\delta_{i,j}$. In this configuration the dipole-dipole interactions are strongly frustrated and lead, for $g = 0$, to large fluctuations (in $S_x$) even in the ordered ground states at $J/\omega_d > 0$. As shown in the corresponding full phase diagram in Fig. 4(a), under this condition completely new cavity-induced phases appear at sufficiently large $g/\omega_c$.

To understand these observations, let us first summarize the established results of the frustrated TFI model at vanishing coupling $g = 0$. In the classical limit $J/\omega_d \to \infty$, the strong frustration prevents the spins from ordering even at zero temperature and the model exhibits an exponentially large (in $N$) ground-state manifold with a finite $T = 0$ entropy density [40]. However, quantum fluctuations from a transverse field, i.e. the term $\omega_d S_x$ in Eq. (1), select an ordered subset of states in an “order-by-disorder” (OBD) process [41]. As shown in the inset in Fig. 4(a), the selected three-sublattice (3SL) antiferroelectric state [42, 43] can be depicted as an arrangement of anti-aligned dipoles on two of the sublattices (in the $S_x$ direction), while on the third sublattice the dipoles align (paraelectrically) with the transverse field and do not point in any particular direction according to $S_x$. This phase is thus characterized by a long-range 3SL order, while it still exhibits strong fluctuations in $S_x$ [see Fig. 4(b), left panel]. When the interaction strength $J/\omega_d$ is decreased below a critical value, the 3SL phase eventually becomes unstable towards a disordered, paraelectric phase. The phase transition is continuous and features an emergent $O(2)$ symmetry, such that the universality class is not of Ising, but of XY type [42, 43].

To investigate the properties of this system for $g/\omega_c > 0$, we compute the correlations corresponding to 3SL order, $\langle \sigma_x \sigma_x^\dagger \rangle^{3SL}$ [see Methods], and find an extended region above a critical line $J^*(g)/\omega_d$ where they become large. This indicates the stability of the 3SL phase also in the presence of the cavity mode [see Fig. 4(a)]. Similar to the square lattice case, increasing the coupling to the cavity reduces the critical value $J^*(g)/\omega_d$, which we estimate by maxima in the order parameter fluctuations $(\Delta p^{3SL})$. For $J < J^*(g)$ we observe the crossover between the paraelectric and the collective subradiant phase also for the triangular lattice, since the geometry becomes irrelevant in this regime. While the formation of such a homogeneous state is hindered by the 3SL order above the transition line $J > J^*(g)$, we discover a new type of ‘3SL subradiant’ regime for $g/\omega_c \gtrsim 3$. This regime is characterized by a finite order, $\langle \sigma_x \sigma_x^\dagger \rangle^{3SL} > 0$, and is thus separated from the collective phase by a sharp
transition line [see Fig. 4(a)]. At the same time it differs from the normal 3SL phase in terms of its vanishing photon number, $\langle a^\dagger a \rangle \simeq 0$, which indicates strongly reduced polarization fluctuations. This difference can be clearly seen in the ground state distribution of $S_x$-values in the two regimes, as shown in Fig. 4(b). While the polarization distribution is broad in the normal 3SL phase, it is pinned to a single value of $S_x = 0$ deep in the subradiant regime. This behavior can be intuitively explained, by adopting again the simplified picture of a 3SL state, where the fluctuating dipoles on one sublattice participate in the formation of a collective subradiant configuration, similar to the state $|\psi_{cs}\rangle$, while the two $S_x$-polarized sublattices remain unaffected. Note, however, that this is only an oversimplified picture of the actual state, where correlations among different sublattices do not vanish completely.

**ORDER BY CAVITY-INDUCED DISORDER**

A very surprising finding in the case of a triangular lattice is the appearance of an additional superradiant phase [blue region in Fig. 4(a)]. As shown by the histogram in Fig. 4(b), also in this phase the polarization is well-defined, but assumes non-zero values $S_x = \pm 1, 2, \ldots$, and consequently, $\langle a^\dagger a \rangle = (g/\omega_c S_x)^2 \gg 1$. Although this value is much smaller than the value in the superradiant phase, this property is completely unexpected, since, at first sight, in this regime both the direct dipole-dipole as well as the cavity-induced interactions would favor a fully anti-aligned configuration. As shown in Fig. 4(c) the transition into this phase is associated with a sharp peak in the photon-number fluctuations, $\langle \Delta a^\dagger a \rangle$, which also remain finite within this phase.

To investigate the properties of this new type of superradiant states we focus on the relevant regime $g/\omega_c \gg 1$, where we can eliminate the photons using strong-coupling perturbation theory. The remaining low-energy physics of $H_{\text{QED}}$ is then approximately described by the effective spin model [2, 21]

$$H_S = \sum_{i<j} J_{ij} \sigma^z_i \sigma^z_j + h_z S_z - J_c (S^2 - S^2_z),$$

where $S = (S_x, S_y, S_z)$. Here, $h_z = \omega_d \exp(-g^2/(2\omega_c^2))$ is the renormalized ‘transverse field’ and $J_c = \omega^2_d \omega_c/(2\omega_c^2) \geq 0$ is the strength of the cavity-mediated collective coupling [see Methods]. Although Eq. (2) is derived under the assumption $J_{ij} \to 0$, a comparison with full numerical simulations up to $N = 24$ shows that $H_S$ accurately reproduces the qualitative features of $H_{\text{QED}}$ for large $J_{ij}$, as long as $g/\omega_c \gtrsim 3$ [see Fig. 4(a)]. As already discussed above, the regular OBD process on the frustrated, antiferroelectric Ising interactions (for $g/\omega_c \lesssim 3$) is driven by the perturbation with a transverse field $\propto S_z$, which stabilizes the normal 3SL phase. On top of that, the collective term $\propto J_c$ can induce a crossover into the 3SL subradiant regime. The appearance of a superradiant phase suggests that this hierarchy no longer holds for large $J$ and $g/\omega_c \gtrsim 3$, where the transverse field $h_z$ is already strongly suppressed. Instead, a new OBD process occurs, where the quantum fluctuations from the cavity-induced collective term select a distinctly ordered subset of states.

A common way to analyze different ordering patterns on the triangular lattice is to consider the polarizations $p_{A,B,C}$ of each sublattice separately and look at the distribution of the 3SL order parameter $p^{3\text{SL}} = p_A + p_B e^{-14\pi/3} + p_C e^{14\pi/3}$ in the complex plane [see Supplementary Section III]. In Fig. 5(a) we use this method to represent the different ground-state structures in the normal, the subradiant and the superradiant 3SL phases. The large values of $\langle |p^{3\text{SL}}| \rangle$ show that all three phases exhibit a large 3SL order, but different levels of fluctuations. Further, the pattern for the superradiant phase differs qualitatively from the other two plots, in particular the positions of the largest peaks are shifted, and indicate a configuration where dipoles in one sublattice
are (almost) fully polarized in $S_z$, while dipoles on the other two sublattices are equally, but only partially polarized along the opposite direction. For $N = 24$ this ordering leaves a residual net polarization $|S_z| = 1$.

Within the effective spin model we can investigate the superradiant phase also for larger lattices and find that this residual polarization increases with the system size and leads, with increasing ratio $J/J_c$, to a whole series of superradiant phases, characterized by $|S_z| = 1, 2, 3, \ldots, S_z^{\max}$ [see Fig. 5(b)]. The maximum value $S_z^{\max}$ obtained in the OBD limit $J_c/J \rightarrow 0$ can be calculated from first order degenerate perturbation theory [see Methods] and is plotted in Fig. 5(c) for different (regular) triangular clusters of up to $N = 48$ sites. From this analysis we can extract a linear scaling for the maximal ground state polarization $S_z^{\max} = \delta N$ and the photon number $\langle a^\dagger a \rangle = (g \delta N/\omega_c)^2$ with a polarization density of $\delta \approx 0.07$. Interestingly, very similar distributions of $p_{3SL}$ and a finite net polarization (although at a much smaller value of $\delta$) have been previously discussed in connection with supersolidity in frustrated spin systems [44–48], where magnetic and superfluid order parameters coexist. While outside the scope of the current study, this connection between superradiance and supersolidity in cavity QED is a particularly exciting direction to explore further.

CONCLUSIONS

In summary, we have addressed the many-body problem in cavity QED, which arises from the interplay between short-range electrostatic interactions and the non-perturbative coupling to a common cavity mode. Based on exact numerical calculations, we have found several novel phases, which have no direct counterpart in the collective Dicke-type models [23, 24, 27] usually studied in quantum optics, nor in regular solid-state spin systems. The basic mechanisms identified in this work, i.e. the cavity-induced reduction of fluctuations, extended superradiant states without order, or the formation of superradiant states through a cavity-induced OBD process, serve as an important guideline to explore similar phenomena also in other types of strongly interacting systems [30–32]. While the most interesting regime $g/\omega_c > 1$ is not accessible in cavity QED experiments with atoms and molecules today, large-scale systems of superconducting two-level systems coupled directly and via microwave modes provide a natural platform to explore this new physical regime. Such systems are currently developed for quantum simulation and quantum annealing schemes [49], where ultrastrong coupling effects, similar to what we have analyzed here, can find direct practical applications [50].

METHODS

Numerical Simulations The numerical results in this manuscript have been achieved by Exact Diagonalization using a Lanczos algorithm [51, 52] on clusters with a finite number of $N$ two-level systems [see Supplementary Section I]. To reduce finite-size effects we use periodic boundary conditions along both directions of the square and triangular lattices. The Hilbertspace $\mathcal{H}$ is kept finite by introducing a photon-number cutoff $n_{\text{ph}}^{\max}$ for the cavity mode in $H_{\text{cQED}}$, such that $a^\dagger |n_{\text{ph}}^{\max}\rangle \equiv 0$ and $\dim(\mathcal{H}) = 2^N (n_{\text{ph}}^{\max} + 1)$. $n_{\text{ph}}^{\max}$ has to be chosen large enough to achieve accurate results throughout the different regimes of the external parameters [see Supplementary Section II]. To further reduce the Hilbertspace dimension, we use the $\mathbb{Z}_2$ symmetry of $H_{\text{cQED}}$, given by the operator $S = e^{-i\pi (x^2 + S_z)}$, together with the lattice translational and point-group symmetries to block-diagonalize $\mathcal{H}$.

On finite systems, symmetries cannot be broken spontaneously and the corresponding local order parameters, such as the total polarization

$$p = S_z = \sum_i \sigma_i^z/2,$$

are strictly zero. However, the correlations $\langle \sigma_i^x \sigma_j^x \rangle$ between spins can be used to identify ordered phases and their ordering patterns. From the spin-spin correlations, we define the (normalized) structure factor

$$\Sigma_x(\vec{k}) = \frac{1}{N} \sum_{i=0}^{N-1} e^{-i\vec{k} \cdot \vec{r}_i} \langle \sigma_i^x \sigma_i^0 \rangle$$

for a momentum $\vec{k}$ in the Brillouin zone of the lattice. Here, $\vec{r}_i = \vec{r}_i - \vec{r}_0$, and we use the translational symmetry to only consider a single reference spin at coordinate $\vec{r}_0$. In an ordered phase $\Sigma_x(\vec{k})$ shows large peaks at specific momenta $\vec{k}^*$, and the value of $\Sigma_x(\vec{k}^*)$ can be used to define the strength of the ordering.

The ferroelectric phase is identified with an ordering momentum $\vec{k}^* = \Gamma \equiv (0, 0)$ and we define $\langle \sigma_x \sigma_x \rangle^{\text{ferro}} = \Sigma_x(\Gamma)$. The Néel (3SL) antiferroelectric phases on the square (triangular) lattice show peaks at $\Gamma \equiv (\pi, \pi)$ ($K \equiv (\pm\pi/3, 0)$) so that we can define the staggered correlations $\langle \sigma_x \sigma_x \rangle^{\text{mag}} = \Sigma_x(M)$ ($\langle \sigma_x \sigma_x \rangle^{3SL} = \Sigma_x(K)$).

To characterize phases with staggered spin patterns it is also convenient to decompose the lattice into sublattices, such that the spins within a sublattice are identical. For the Néel phase, the square lattice can be decomposed into two sublattices ($A$, $B$), and we can decompose the triangular lattice into three sublattices ($A$, $B$, $C$) to match the staggered correlation pattern. Then, defining the sublattice polarizations $p_I = \sum_{i \in I} \sigma_i^z/2$, the order
parameter correlations can be computed from
\[
\langle \sigma_x \sigma_x \rangle_{\text{ferro}}^{\text{ferro}} = \frac{4}{N} \left| \langle p \rangle \right|^2 \\
\langle \sigma_x \sigma_x \rangle_{\text{stag}} = \frac{4}{N} \left| \langle p_A - p_B \rangle \right|^2 \\
\langle \sigma_x \sigma_x \rangle_{\text{3SL}}^{\text{3SL}} = \frac{4}{N} \left| \langle p_A + p_B e^{-i4\pi/3} + p_C e^{i4\pi/3} \rangle \right|^2.
\]
(5)

This formulation also illustrates the relation of these correlation measures to the corresponding order parameters
\[
p_{\text{stag}} = p_A - p_B \\
p_{\text{3SL}} = p_A + p_B e^{-i4\pi/3} + p_C e^{i4\pi/3}.
\]
(6)

To track finite-size phase boundaries we introduce the fluctuations
\[
\langle \Delta O \rangle = \langle O^2 \rangle - \langle O \rangle^2
\]
(7)
of an observable $O$ and observe that $\langle \Delta |O| \rangle$ obeys a peak at the transition between the disordered phase and the phase ordered according to the order parameter $O$.

To distinguish different 3SL patterns on the triangular lattice, we decompose the complex 3SL order parameter as
\[
p_{3\text{SL}} = |p_{3\text{SL}}| e^{i\theta},
\]
(8)
where $|p_{3\text{SL}}|$ describes the strength of the 3SL order, while the angle $\theta$ identifies different ordering patterns. In this manuscript, we have observed two distinct 3SL patterns which we can define by the sublattice polarizations $\vec{p} = (p_A, p_B, p_C)$. As described in the main text, for the 3SL normal and subradiant regimes, we find $\vec{p}^{N/\text{Sub}} = (0, m, -m)$, with $m > 0$. In the superradiant phase, on the other hand, the pattern $\vec{p}^{\text{Sup}} = (-m, n, n)$ with $n \geq m/2$ is stabilized. These different classical 3SL patterns can be distinguished by evaluating the distribution of $p_{3\text{SL}}$ in the complex plane, which shows six peaks at distinct angles $\theta$ [see Supplementary Section III]. The full quantum-mechanical ground states then obey additional fluctuations around the classical configurations, as shown in Fig. 5(a), which can be further used to distinguish the normal and subradiant 3SL regimes.

### Polaron Transformation & Effective Spin Model

In the ultrastrong coupling regime it can be convenient to transform $H_{\text{cQED}}$ [Eq. (1)] into a frame of displaced photon number states (polarons) by applying the unitary operator $U = e^{g/\omega_c S_z (a^\dagger - a)}$. The Hamiltonian $H_{\text{cQED}}$ transforms into
\[
\tilde{H}_{\text{cQED}} = U H_{\text{cQED}} U^\dagger = \omega_c a^\dagger a + \sum_{i<j} \frac{J_{ij}}{4} \sigma_x^i \sigma_x^j + \omega_d U S_z U^\dagger,
\]
(9)
since $a^\dagger \rightarrow a^\dagger - g/\omega_c S_z$ is displaced proportional to $S_z$. Within this formulation, it becomes obvious that the correct electrostatic limit $H_{\text{cQED}} \approx \omega_c a^\dagger a + \sum_{i<j} J_{ij} \sigma_x^i \sigma_x^j$ is achieved for $\omega_d \rightarrow 0$, since the dephasing shift in Eq. (1) exactly cancels the additional terms $\propto S_z^2$ from the transformation of $a^\dagger a$.

The Hamiltonian Eq. (9) can also be advantageous to study superradiant phases, because the photon number $\langle a^\dagger a \rangle$ in this polaron frame ignores the part from the direct coupling to the polarization $S_z$ and remains much smaller than in the standard frame, in particular for superradiant phases. Therefore, a substantially lower photon number cutoff $n_{\text{ph}}^{\max}$ can be sufficient for precise numerical simulations, with the disadvantage of having to deal with a dense photonic Hamiltonian, when $\omega_d \neq 0$.

Also, the polaron photon number, which can be computed by $\langle a^\dagger a \rangle_{\text{polaron}} = \langle (a^\dagger - a)(a - a^\dagger) \rangle$ with $a = g/\omega_c S_z$ in the standard frame, can be a useful observable. In particular, we use characteristic peaks in the polaron photon number to identify the crossover regime between the paraelectric and collective subradiant phases [see Supplementary Section IV].

Using strong-coupling perturbation theory for $g/\omega_c \gg 1$ and projecting onto the lowest-energy sector without polaronic excitations $|0\rangle^\text{pol}_\text{ph}$, the last term in $\tilde{H}_{\text{cQED}}$ can be approximated as $[2,21]$
\[
\omega_d U S_z U^\dagger \approx \omega_d e^{-\frac{x^2}{4\omega_c^2}} S_z - \frac{\omega_d^2 \omega_c^2}{2g^2} (S_z^2 - S_z^4),
\]
(10)
where we have introduced the total spin operator $S = (S_x, S_y, S_z)$. Within this approximation, we thus obtain the effective spin model $H_S \equiv 43$ in the main text. It is important to note, that the eigenstates in the original basis $|\Psi\rangle = e^{g/\omega_c S_z (a^\dagger - a)} |\Psi_{\text{spin}} \otimes |0\rangle^\text{pol}_\text{ph}\rangle$, and the photon number $\langle a^\dagger a \rangle = g^2/\omega_c^2 (\langle S_z \rangle^2)$ is generally non-zero in the standard basis. Note that the approximate expression in Eq. (10) has been derived for non-interacting dipoles $J_{ij} = 0$.

**OBD Simulations** The classical Ising model, which is obtained from $H_{\text{cQED}}$ or $H_S$ for $J \rightarrow \infty$, is strongly frustrated on the triangular lattice with nearest-neighbor interactions and does not order even at zero temperature $T = 0$. It features an exponentially large (in $N$) ground-state manifold, with an extensive $T = 0$ entropy $S \approx 0.332k_B N$ [53]. This ground-state manifold can be destabilized by quantum fluctuations from other interaction terms, such as a transverse field or the cavity-mediated collective coupling $\propto J_c$ in $H_S$, when a particular subset of states, with the softest response to the fluctuations, is selected. When the set of the selected states is ordered, this process is termed “order by disorder” [41], and for a perturbation with a transverse field this mechanism is known to induce a 3SL ordered phase [42, 43].

To study the OBD process from the collective coupling $\propto J_c$ in Eq. (2) in the $J/J_c \rightarrow \infty$ regime, we restrict the
Hilbert space of the system to the degenerate, classical ground-state manifold, and define the effective Hamiltonian

\[ H_{\text{OBD}} = -\mathcal{P}_J (S^2 - S_x^2) \mathcal{P}_J. \]  

Here, \( \mathcal{P}_J \) is the projector onto the classical ground-state manifold. The low-energy eigenvectors of \( H_{\text{OBD}} \) yield the states stabilized by the OBD mechanism (in the sense of a first-order degenerate perturbation theory), from which we can compute the observables, as shown in Fig. 5 in the main text. The advantage of this approach is that, compared to a full simulation of \( H_S \), larger system sizes \( N \) can be simulated.

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**AUTHOR CONTRIBUTIONS**

P.R. proposed the project and M.S. performed all the numerical simulations. All authors contributed to the discussion of the results and the writing of the manuscript.

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SUPPLEMENTARY INFORMATION:
CAVITY QUANTUM ELECTRODYNAMICS OF STRONGLY CORRELATED DIPOLAR MATTER

I. FINITE-SIZE CLUSTERS FOR SIMULATIONS

In this section we present the finite-size clusters with $N$ sites used in the numerical simulations [see Supplementary Fig. 1]. We utilize periodic boundary conditions along two directions of the clusters to reduce finite-size effects. Also, to study genuine two-dimensional properties we only use clusters with an aspect ratio $\epsilon = 1$, i.e. the loops around both periodic directions have equal length. To fit the antiferroelectric phases with a two (three) sublattice structure, we only consider square (triangular) clusters with $N$ mod 2 = 0 ($N$ mod 3 = 0). Here, it is worth mentioning that the subradiant states discussed in this work cannot exist on clusters with odd $N$, where the possible total polarizations $S_x$ are half-integers, so that a subradiant state with fixed $S_x = 0$ cannot be obtained.

Supplementary FIG. 1. Some of the finite-size clusters used in the simulations. The top (bottom) row shows square (triangular) clusters. The black dots are the sites of the finite-size clusters, the yellow background illustrates the Wigner-Seitz cell. The yellow lines indicate the nearest-neighbor bonds.

II. PHOTON NUMBER CUTOFF

In addition to using a finite number of dipoles $N$, we have to introduce a photon number cutoff $n_{\text{ph}}^\text{max}$ to obtain a finite Hilbert space for the numerical simulations. This cutoff has to be chosen large enough such that the true ground state in the full Hilbert space only shows negligible deviations (up to some defined precision) when it is projected into the restricted Hilbert space. Appropriate values for $n_{\text{ph}}^\text{max}$ strongly depend on the chosen external parameters, i.e., for parameters belonging to a superradiant phase much larger cutoffs have to be chosen than for parameters belonging to a subradiant phase.

For the simulations in this work we choose $n_{\text{ph}}^\text{max}$ large enough, such that doubling this cutoff does not change the measured observables. In Supplementary Fig. 2 we show an analysis of the dependence of observables on $n_{\text{ph}}^\text{max}$ for a square lattice configuration with $N = 16$ dipoles and a constant coupling $g/\omega_c = 2$. For antiferroelectric interactions, $J/\omega_c > 0$, a small cutoff $n_{\text{ph}}^\text{max} = 64$ is already sufficient to obtain converged results in all observables, since the average photon number $\langle a^\dagger a \rangle$ remains small. Contrarily, for ferroelectric interactions, $J/\omega_c < 0$, the average photon number and its fluctuations become large [see Supplementary Fig. 2(a)] when the superradiant regime is entered. Then, a too small cutoff yields false results not only for $\langle a^\dagger a \rangle$, but also for pure dipole observables [see Supplementary Fig. 2(c)-(f)]. The distribution of the photon number $a^\dagger a$ in the ground state [see inset in Supplementary Fig. 2(a)] reveals, that a cutoff $n_{\text{ph}}^\text{max} \gtrsim 400$ would be sufficient for a simulation with these particular external parameters.

In Supplementary Fig. 2 we also show results obtained from the polaron frame with Hamiltonian $\tilde{H}_{\text{QED}}$. In this formulation, a much smaller cutoff is already enough to obtain converged results in the ferroelectric regime, since the
Supplementary FIG. 2. Scaling of observables with the photon number cutoff. Different symbols indicate different photon number cutoffs $n_{\text{ph}}^\text{max}$ used in the simulations. The red hexagon shows results of a simulation in the polaron frame, while all other symbols represent simulations in the standard frame. (a) Photon number and (b) polaron photon number, (c-d) order parameter correlations for the (c) ferroelectric, and (d) Néel phase, (e-f) fluctuations of the (e) polarization, (f) staggered polarization. The inset in (a) shows the ground state distribution of the photon number $a^\dagger a$ for $J/\omega_d = -0.5$, as indicated by the vertical dashed line, for $n_{\text{ph}}^\text{max} = 64$ (4096) in blue (green) color.

Polaron photon number $\langle a^\dagger a \rangle^{\text{polaron}}$ remains small even in the superradiant phase [see Supplementary Fig. 2(b)].

III. HISTOGRAMS OF THE THREE-SUBLATTICE ORDER PARAMETER

In Supplementary Fig. 3 we illustrate the properties of 3SL order parameter histograms in the complex plain. A (classical) state with sublattice polarizations $\vec{p} = (p_A, p_B, p_C)$ gives a single peak in the histogram according to the diagram shown in Supplementary Fig. 3(a). While this identification is not unique for any $\vec{p}$, the strength of the 3SL ordering $|p^{\text{3SL}}|$ is given by the distance of the peak from the center, and the angle $\theta$ (in the complex plain) indicates different types of 3SL ordering patterns. In particular, states of type $\vec{p} = (0, 1, -1)$ with two fully, but oppositely polarized, and a non-polarized sublattice (zero net polarization), give peaks at the centers of the hexagonal boundaries of the histogram. The six different peaks correspond to the possible permutations of the three sublattices. States of type $\vec{p} = (1, 1, -1)$, where all sublattices are fully polarized, one of them oppositely to the others (non-zero net polarization) have peaks at the edges of the hexagonal boundary. The six peaks correspond to the possible permutations of the sublattices and the inversion of the polarization $\vec{p} \rightarrow -\vec{p}$.

More generally, patterns of type $\vec{p} = (0, m, -m)$ yield peaks at angles $\theta_l = \pi/6 + l\pi/3$, $l \in \{0, \ldots, 5\}$, with a radius proportional to $m$, as shown in Supplementary Fig. 3(b). A comparison with the full histograms for the normal and 3SL subradiant regimes [c.f. Fig. 5(a) in the main text] shows that the maxima for those phases correspond to such a pattern with maximal $m = 1$, as indicated by the red dots. Furthermore, we want to note that fluctuations of the non-polarized sublattice lead to a broadening of the six single peaks parallel to the edges of the hexagonal boundary, as seen for the normal 3SL phase found in the main text. On the other hand, patterns of type $\vec{p} = (1, -1/2 - \varepsilon, -1/2 - \varepsilon)$ with a ground state polarization $|S_x| = 2\varepsilon N/3$ have peaks at angles $\theta_l = l\pi/3$, $l \in \{0, \ldots, 3\}$, with a large non-zero radius, which depends on $\varepsilon$, as shown in Supplementary Fig. 3(c). The red circles show the positions of the maxima found in the full histograms of the 3SL superradiant phase in the main text, which has a non-zero net polarization $|S_x| = 2$ ($N = 36$) [c.f. Fig. 5(a) in the main text]. We want to note, that all sublattice patterns of type $\vec{p} = (m, -n, -n)$ yield peaks with $\theta_l = l\pi/3$.

IV. ESTIMATING CROSSOVER BOUNDARIES

In contrast to phase transitions with an abrupt change in behavior of the order parameter (in the thermodynamic limit), crossovers between two regimes of states with different physical properties show a smooth change in some
of the observables. Therefore, the crossover region, or a ‘boundary’ between the two regimes, can typically not be determined uniquely, but depends on the chosen observable and the feature used to estimate the boundary.

To estimate a boundary between the paraelectric regime and the collective subradiant regime, we use maxima in the polaron photon number $\langle a^\dagger a \rangle_{\text{polaron}}$, as shown in Supplementary Fig. 4. In comparison, the standard photon number $\langle a^\dagger a \rangle$ is a bad estimator for the crossover when $J/\omega_c < 0$, where the proximity to the superradiant phase spoils its characteristic features in the narrow collective subradiant regime.

Based on this definition, we observe a shift of the boundary to larger $g/\omega_c$ with increasing the system size $N$ [see also Fig. 2(a) in the main text], consistent with the analysis of the collective spin model [21] for $J_{ij} = 0$. The shift of this boundary is too weak to make reliable predictions about the fate of the collective subradiant phase in the limit $N \to \infty$, using exact diagonalization techniques. In this respect it is important to emphasize, that this thermodynamic limit is also not properly defined for the single-mode model used in this work, where $H_cQED$ is super-extensive. For finite systems, this approximation is, however, expected to capture the main results and our analysis can be directly applied to most near-term experiments, where intermediate-scale systems, far away from the thermodynamic limit, will be realized.

Because of the similarity with the evolution from the paraelectric to the collective subradiant regimes, we also expect the evolution from the normal to the subradiant 3SL regime to be described by a crossover instead of a sharp phase boundary. We characterize the regimes by a distinct strength of the polarization fluctuations $\langle \Delta |p| \rangle$, since the polarization fluctuates strongly in the 3SL normal regime and becomes pinned to $S_x = 0$ in the 3SL subradiant regime [see Fig. 4(b) in the main text]. We, therefore, define the boundary by a rather sharp drop in $\langle \Delta |p| \rangle$ and estimate its location for constant $J/\omega_d$ by a peak in the negative gradient of $\langle \Delta |p| \rangle$ with respect to the coupling $g/\omega_c$ [see Supplementary Fig. 5].
Supplementary FIG. 4. Crossover between paraelectric and collective subradiant regimes. Open (filled) symbols show the polaron photon number on a $N = 16$ ($N = 26$)-sites square lattice for constant values of $J/\omega_d$. We use the peak positions, indicated by the dashed vertical lines for $N = 26$, to estimate the crossover boundary between the paraelectric (Para) and the collective subradiant (CS) regimes [c.f. Fig. 2(a) and Fig. 4(a) in the main text].

Supplementary FIG. 5. Crossover between the normal 3SL and subradiant 3SL regimes. The polarization fluctuations (left panel) and their gradient with respect to $g/\omega_c$ (right panel) are shown for constant $J/\omega_d = 1$. The crossover boundary is estimated by a sharp drop in the fluctuations $\langle \Delta |p| \rangle$, where the polarization distribution becomes strongly pinned to the single value $S_x = 0$. As shown in the right panel, we compute the boundary position by a peak in the negative gradient of $\langle \Delta |p| \rangle$ with respect to the external parameter $g/\omega_c$ (indicated by the dashed vertical line).