Uniqueness of the phase transition in many-dipole systems

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Due to various no-go and counter no-go results, the existence and natures of thermodynamic phases of light-matter systems continue to be disputed. We show using an arbitrary-gauge approach that a unique phase transition does occur in many-dipole cavity QED systems, and manifests unambiguously via a macroscopic gauge-invariant polarisation. Whether the abnormal phase appears superradiant depends on the gauge, because the gauge controls the extent to which the polarisation is included as part of the radiative quantum subsystem. A separate issue is that each gauge provides a different two-level approximation of the material dipoles. Our approach enables characterisation of physical behaviour in terms of any chosen material and radiation subsystems, using any of the non-equivalent approximate models. We provide numerical results for a finite number of dipoles without the two-level approximation, exhibiting clear precursors to the phase transition and demonstrating how accurate approximate predictions can be identified. Arbitrary-gauge QED therefore eliminates any apparent inconsistencies in the description of many-dipole cavity QED systems and their thermodynamic phases.

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I. INTRODUCTION

Superradiance was originally described by Dicke [1], and since then it has received a great deal of attention (see for example [2] for a recent introduction). A superradiant phase of a light-matter system is one in which a macroscopic number of photons arises due to the interaction between many dipoles. The possibility of such a phase transition within the Dicke model was first acknowledged some time ago [3, 4]. Later, seminal contributions were made in the connection with quantum chaos using the Holstein-Primakoff mapping [5–7]. The topic now includes extended Dicke models [8–12], driven and open systems, semi-classical descriptions [13–17], and artificial QED systems [18–29].

One of the most controversial aspects of theoretical studies on superradiance has been the validity of the so-called “no-go theorem” [30]. This theorem requires that the often neglected diamagnetic $A^2$-term of the Coulomb gauge is retained. It has been both refuted and confirmed in numerous subsequent works [11, 12, 18, 21, 25, 31–40]. Notably, a circumvention of the no-go theorem has been thought to require artificial light-matter systems involving, for example, specific superconducting qubits [21, 27]. As well as the $A^2$-term the role of electrostatic interactions has also been recognised as important [19, 31, 33, 34, 36, 37]. In fact, permutations of no-go and counter no-go results that consider different Hamiltonian interaction terms can be found within the literature. In the multipolar gauge direct inter-atomic electrostatic interactions and the $A^2$-term are both absent, so the superradiant phase transition appears to be recoverable [37, 38]. If in a multipolar gauge formulation explicit dipole-dipole interactions that are not naturally present, are added in, then a no-go theorem emerges without the presence of the $A^2$-term [19, 34, 41, 42]. On the other hand, if both dipole-dipole interactions and the $A^2$-term are retained within the Coulomb-gauge treatment, then an apparently different ferroelectric phase transition is predicted to occur [36]. The occurrence or otherwise of a phase transition, its cause, and its physical nature, appear to depend on the choice of gauge, which determines the interaction terms that are explicit within the Hamiltonian.

A related issue is the breakdown of gauge-invariance due to the two-level truncation of material dipoles [41–43]. In Ref. [29] a minimal working model of a hybrid many-dipole $LC$-resonator system was constructed based on the inclusion of dipole-dipole interactions within the equations of motion. The two-level truncation was found to cause gauge non-invariance. It was also found that a ferroelectric instability occurs due to dipole-dipole interactions, and that this can lead to superradiance of the corresponding radiation mode (the $LC$-oscillator). This superradiance due to electrostatic interactions was noted to contrast with the usually held view in which it arises due to the coupling to radiation. On the other hand, Keeling has considered a traditional cavity QED system, and found that a ferroelectric phase transition does occur in the Coulomb gauge, but that this should not incur any macroscopic occupation of the corresponding radiation mode [36].

The occurrence and nature of phase transitions in many-dipole light-matter systems, and how this relates to the choice of gauge remains unclear. A basic ingredient that is lacking is a fundamental derivation of the correct underlying Hamiltonian together with a self-consistent use of all valid approximations, but without the commitment to particular fixed gauges. This gap will be filled.

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in the present work. Our first-principles derivation starts with the basic Maxwell-Lorentz theory and is carried out in an arbitrary gauge parameterised by a real number \( \alpha \). We carry out the single-mode approximation in such a way as to preserve not only gauge-invariance, but also the fundamental algebraic and kinematic relations of the theory.

Up to the point of truncating the material dipoles to two levels each, our treatment is fully gauge-invariant. At this stage predictions for any given physical observable could not possibly depend on which interaction terms are present through a particular choice of gauge. Even upon making the two-level approximation, we are able to prove that a unique physical phase transition does generally occur in generic many-dipole cavity QED systems. Thus, the original “no-go theorem” that has prompted extensive debate in the past, is in the first place physically baseless even when only considering cavity QED systems. We show that the abnormal phase of the system is unambiguously signalled by a macroscopic average of the gauge-invariant transverse polarisation field \( P_T \), which equals the longitudinal electric field \( E_L \) except at the point-dipole positions themselves. Whether the abnormal phase is characterised as ferroelectric or as superradiant depends on the choice of gauge, which controls the relative extent to which the field \( P_T \) is included within the reactive quantum subsystem. Importantly, our approach clearly shows that different viewpoints provided by different gauges are not contradictory, they are in fact equivalent. Specifically, they are simply different ways of viewing the same phenomenon in terms of physically different light and matter subsystems.

Despite the resolutions provided to the outstanding issues mentioned, the two-level approximation is found to break the gauge-invariance of the theory. Thus, although our results eliminate any ambiguity regarding the occurrence of a unique phase transition, as well as all qualitative ambiguities regarding both its cause and its physical manifestation, some quantitative ambiguity inevitably remains within the approximate description. This cannot be resolved without a comparison with the fully gauge-invariant non-truncated theory. Although such a comparison has been performed in the single-dipole case \([41– 43]\), the many-dipole case is more formidable. For the specific example of a collection of \( N \) dipoles each having a double-well potential, we provide numerical results for up to \( N = 4 \), avoiding the two-level approximation. These results clearly exhibit precursors to the phase transition and show how accurate two-level model predictions can be identified. For any example in which there is a two-level truncation that remains accurate as \( N \) finitely increases, it is reasonable to conclude that the same two-level truncation will be accurate in the thermodynamic limit \( N \to \infty \).

In summary, the arbitrary-gauge approach to QED is used to eliminate all ambiguity surrounding the qualitative predictions of the phase transition in many-dipole systems, including its underlying cause and its macroscopic manifestation. It is also shown to be capable of eliminating any further quantitative ambiguity resulting from the use of material two-level truncation.

II. SINGLE-MODE \( N \)-DIPOLE HAMILTONIAN

We adopt a general formulation of QED in which the gauge is selected by choosing the value of a real parameter \( \alpha \). We consider \( N \) identical hydrogen-like atoms within the electric-dipole approximation (EDA). Each dipole has a reduced mass \( m \) and charge \(-e\). The \( \mu \)th dipole is described by a (classical) centre-of-mass position \( R_\mu \) and a quantum relative position operator \( r_\mu \), which in turn defines a dipole moment operator \( \mathbf{d}_\mu = -e \mathbf{r}_\mu \). The dipoles interact with a common electromagnetic field described by transverse-electric and magnetic fields \( E_T \) and \( B \) respectively. The Hamiltonian for the system is derived from first principles in Supplementary Note 1 and can be written in the gauge-invariant form \([43]\)

\[
H = E_{\text{matter}} + E_{\text{field}}
\]

where

\[
E_{\text{matter}} := \sum_{\mu=1}^{N} \frac{1}{2} m \dot{r}_\mu^2 + V + V_{\text{dip}},
\]

\[
E_{\text{field}} := \frac{1}{2} \int d^3x \left[ E_T(x)^2 + B(x)^2 \right].
\]

Here \( V \) denotes the total intra-dipole potential, and \( V_{\text{dip}} \) denotes the inter-dipole electrostatic energy. The \( \alpha \)-dependent canonical momenta are found to be (See Supplementary Note 1)

\[
P_{\mu \alpha} = m \dot{r}_\mu - e(1 - \alpha) \mathbf{A}(R_\mu),
\]

\[
\Pi_\alpha(x) = -E_T(x) - P_{T \alpha}(x),
\]

where \( \mathbf{A} \) is the gauge-invariant transverse vector potential such that \( E_T = -\mathbf{A} \) and \( P_{T \alpha} \) is the \( \alpha \)-gauge material transverse polarisation given by

\[
P_{T \alpha}(x) = \alpha P_T(x),
\]

\[
P_{T, i}(x) = \sum_{\mu=1}^{N} \frac{\delta_{\mu, j}}{\mu} (x - R_\mu).
\]

Here, and throughout, we assume the Einstein summation convention for repeated Latin indices. In Eqs. (6) and (7) \( P_T \) is a gauge-invariant transverse polarisation field (in EDA), which coincides with \( P_{T \alpha} \) in the multipolar-gauge \( (\alpha = 1) \). Canonical operators belonging to distinct gauges are related by a unitary gauge-fixing transformation defined in terms of \( P_{T \alpha} \) (see Supplementary Note 1).

We now restrict our attention to a single radiation mode within a cavity of volume \( v \), wave-vector \( \mathbf{k} \) and unit polarisation vector \( \mathbf{\epsilon} \) such that \( \mathbf{k} \cdot \mathbf{\epsilon} = 0 \) and \( |\mathbf{k}| = \omega \).
is the frequency of the mode. The mode is described by bosonic operators $a_\alpha$, $a_\alpha^\dagger$ with $[a_\alpha, a_\alpha^\dagger] = 1$. The details of the modification of the Hamiltonian resulting from the single-mode restriction are given in Supplementary Note 1. The single-mode restriction is imposed in Fourier-space consistently on all fields, including the transverse delta-function $\delta^T$. This eliminates the need to regularise the single-dipole polarisation self-energy contributions ($P^2_{T\mu}$-terms) [44], and ensures that the fundamental transverse canonical commutation relation $[A_{T,i}(x), \Pi_{T,j}(x')] = \delta^T_{ij} (x-x')$ is preserved. The fundamental kinematic relations given by Eqs. (4) and (5), are also preserved.

In order to obtain a Dicke Hamiltonian we must first take the limit of closely spaced dipoles around the origin $\mathbf{0}$ such that $R_\mu \approx \mathbf{0}$. Details are given in Supplementary Note 1. The final result is the following Hamiltonian

$$H = \sum_{\mu=1}^N \frac{1}{2m}(\mathbf{p}_\mu + e(1-\alpha)\mathbf{A})^2 + V + \frac{\alpha^2}{2\nu} \sum_{\mu=1}^N (\mathbf{d}_\mu \cdot \mathbf{e})^2$$

$$+ (1-\alpha^2)V_{\text{dip}} + \alpha \sum_{\mu=1}^N \mathbf{d}_\mu \cdot \Pi_\alpha + \omega \left(a_\alpha^\dagger a_\alpha + \frac{1}{2}\right)$$

(8)

where

$$\mathbf{A} = \frac{\mathbf{e}}{\sqrt{2\omega \nu}}(a_\alpha^\dagger + a_\alpha),$$

(9)

$$\Pi_\alpha = i\varepsilon \sqrt{\frac{\omega}{2\nu}} (a_\alpha^\dagger - a_\alpha),$$

(10)

and

$$V_{\text{dip}} = -\frac{1}{2\nu} \sum_{\mu \neq \nu}^N (\mathbf{d}_\mu \cdot \mathbf{e})(\mathbf{d}_\nu \cdot \mathbf{e}),$$

(11)

$$P_{T\alpha} = \frac{\alpha}{\nu} \sum_{\mu=1}^N \varepsilon (\mathbf{d}_\mu \cdot \mathbf{e}).$$

(12)

The theory remains gauge-invariant, because gauge transformations remain unitary. The canonical momenta of different gauges are related via $p_\alpha = R_{\alpha\alpha'} p_{\alpha'} R_{\alpha\alpha}$ and $\Pi_\alpha = R_{\alpha\alpha'} \Pi_{\alpha'} R_{\alpha\alpha}$ where

$$R_{\alpha\alpha'} = \exp \left[i(\alpha - \alpha') \sum_{\mu=1}^N \mathbf{d}_\mu \cdot \mathbf{A} \right]$$

(13)

is the final form of the unitary gauge-fixing transformation between the $\alpha$ and $\alpha'$ gauges.

Note that the usual starting point for the derivation of the “no-go theorem” for a superradiant phase in the (Coulomb-gauge) Dicke model sets $\alpha = 0$ in Eq. (8), but ignores $V_{\text{dip}}$, and is therefore inconsistent. As well as the electric dipole and single-mode approximations, to obtain a Dicke Hamiltonian one must assume that the dipoles themselves are close enough that they can be approximated as interacting with the transverse vector potential at the same point $\mathbf{0}$. Typical inter-atomic separations must therefore be small compared with the wavelength of the radiative mode. If this is the case then inter-atomic dipole-dipole interactions are certainly not negligible. We will show in what follows that the standard “no-go theorem” for a phase transition [30] is physically baseless, because it is not obtained in any gauge if a consistent treatment of interactions is adopted.

We note that there is also a lower bound on the typical inter-atomic separation. If the atoms are sufficiently dense that orbital overlap occurs then the assumption of well-localised separate dipoles would not seem to be valid. For such atomic densities an extended Dicke-model would be required [40]. The provision of such a treatment within an arbitrary-gauge should be straightforward, but since the present simplifying assumption of localised separate dipoles does not affect gauge-invariance, for simplicity, such extended Dicke models are not considered here.

III. NON-EQUIVALENT DICKE MODELS

A. Derivation of the arbitrary-gauge Dicke Hamiltonian

All that remains to obtain a Dicke model from Eq. (8) is to approximate the dipoles as two-level systems. We therefore begin by implementing the approximation

$$H^\alpha_m = \sum_{\mu=1}^N \frac{P^2_{T\mu}}{2m} + V = \sum_{\mu=1}^N \sum_{n=0}^\infty \epsilon_n \langle \epsilon^\alpha_{\mu,n} | \epsilon^\alpha_{\mu,n} \rangle$$

$$\approx \omega_m \sum_{\mu=1}^N \sigma^z_{\mu} + \frac{N}{2} (\epsilon_0 + \epsilon_1)$$

(14)

where the approximate equality results from truncating the sum over dipole levels $n$ at the second level. Here $\sigma^z_{\mu} = [\sigma^+_{\mu}, \sigma^-_{\mu}]/2$ where $\sigma^+_{\mu} = |\epsilon^\alpha_{\mu,1}\rangle \langle \epsilon^\alpha_{\mu,0}|$ and $\sigma^-_{\mu} = |\epsilon^\alpha_{\mu,0}\rangle \langle \epsilon^\alpha_{\mu,1}|$ are raising and lowering operators for the first two levels of the $\mu$th dipole. The $\epsilon_{\alpha}$ are the eigenenergies of a single dipole. They are $\mu$-independent, because the dipoles are assumed to be identical, and they are $\alpha$-independent, because $H^\alpha_m = R_{\alpha\alpha'} H^\alpha_{m'} R_{\alpha\alpha'}$. The material transition frequency is $\omega_m = \epsilon_1 - \epsilon_0$. The eigenstates $|\epsilon^\alpha_{\mu,n}\rangle$ of each dipole are $\alpha$-dependent due to the $\alpha$-dependence of the single-dipole energy operators. We note at this point that various alternative conventions for the implementation of the two-level approximation are available. We discuss these alternatives and provide justification for the chosen conventions in the following section IIIIB.

The total projection onto the first two levels of every dipole is

$$P_\alpha = \bigotimes_{\mu=1}^N \left[ |\epsilon^\alpha_{\mu,0}\rangle \langle \epsilon^\alpha_{\mu,0}| + |\epsilon^\alpha_{\mu,1}\rangle \langle \epsilon^\alpha_{\mu,1}| \right],$$

(15)
such that the right-hand-side of Eq. (14) can be written $P_a H_m^{\alpha} P_a$. Similarly, in the two-level approximation the material canonical operators are taken to be $P_a \hat{d}_\mu P_a = \hat{d}(\sigma_{\mu}^+ + \sigma_{\mu}^-)$ and $P_a (-e p_{\mu a}/m) P_a = i \omega_m \hat{d}(\sigma_{\mu}^+ - \sigma_{\mu}^-)$ where the transition dipole moment $\hat{d} = (\epsilon_{\mu,0}) \hat{d}_{\mu a} | \epsilon_{\mu,1} \rangle$ is assumed to be real and is $\mu$-independent (identical dipoles). If we now introduce the collective operators

$$J^i_\alpha = \sum_{\mu=1}^{N} \sigma_{\mu a}^i, \quad i = \pm, \tau,$$

then the two-level approximation can be taken to constitute the replacements

$$H_{m,\alpha} \rightarrow \omega_m J^z_\alpha + \frac{N}{2} (\epsilon_0 + \epsilon_1),$$

$$\sum_{\mu=1}^{N} \hat{d}_\mu \rightarrow \hat{d}(J^+_\alpha + J^-_\alpha),$$

$$\frac{e}{m} \sum_{\mu=1}^{N} p_{\mu a} \rightarrow -i \omega_m \hat{d}(J^+_\alpha - J^-_\alpha).$$

We also introduce new cavity bosonic operators $c_\alpha$ and $c_\alpha^\dagger$ defined such that

$$\omega \left( a_\alpha^\dagger a_\alpha + \frac{1}{2} \right) + \frac{e^2}{2\mu} (1 - \alpha)^2 N A^2 = \omega_\alpha \left( c_\alpha^\dagger c_\alpha + \frac{1}{2} \right),$$

where

$$\omega_\alpha^2 = \omega^2 + \frac{e^2}{m} (1 - \alpha)^2 \rho.$$

There is no $A^{2n}$-term within the multipolar gauge $\alpha = 1$, implying that $c_1 \equiv c_\alpha$ and $\omega_1 \equiv \omega$.

With the definitions in Eqs. (17)-(21) the Hamiltonian in Eq. (8) becomes an $\alpha$-gauge Dicke Hamiltonian written in terms of collective operators and a renormalised mode:

$$H_{m,\alpha}^{\alpha,2} = \omega_m J^z_\alpha + \frac{N}{2} (\epsilon_0 + \epsilon_1) + \frac{1}{2} \rho d^2 + \omega_\alpha \left( c_\alpha^\dagger c_\alpha + \frac{1}{2} \right)$$

$$- \frac{C_\alpha}{N} (J^+_\alpha + J^-_\alpha)^2$$

$$- i \frac{g_\alpha}{\sqrt{N}} (J^+_\alpha - J^-_\alpha) (c_\alpha^\dagger + c_\alpha)$$

$$+ i \frac{g_\alpha}{\sqrt{N}} (J^+_\alpha + J^-_\alpha) (c_\alpha^\dagger - c_\alpha)$$

where

$$C_\alpha := \frac{1}{2} \rho d^2 (1 - \alpha^2),$$

$$g'_\alpha := (1 - \alpha) \omega_m d \sqrt{\frac{\rho}{2\omega_\alpha}},$$

$$g_\alpha := \alpha d \sqrt{\frac{\rho \omega_\alpha}{2}}.$$

Here $d := \epsilon \cdot d$ and $\rho = N/v$, which remains finite in the thermodynamic limit $N \rightarrow \infty$, $v \rightarrow \infty$. The parameters $C_\alpha$, $g'_\alpha$ and $g_\alpha$ respectively denote the $\alpha$-gauge’s electrostatic coupling strength, Coulomb-gauge-like radiative coupling strength, and multipolar-gauge-like radiative coupling strength. Although the non-truncated Hamiltonian $H$ is unique, we now have a continuous infinity of Dicke Hamiltonians $H_{m,\alpha}^{\alpha,2}$ such that $H_{m,\alpha}^{\alpha,2}$ and $H_{m,\alpha'}^{\alpha,2}$ are not equal when $\alpha \neq \alpha'$. Thus, the two-level truncation of the dipoles has broken the gauge-invariance of the theory [41–43].

### B. Other ways to implement the two-level approximation

Before proceeding to apply the $\alpha$-gauge Dicke Hamiltonian in Eq. (22) we note that the above implementation of the two-level truncation is not unique. One reason for this is that for any operator $O(\hat{d}_\mu, p_{\mu a})$, which is not linear in the material canonical operators we have [43]

$$P_a O(\hat{d}_\mu, p_{\mu a}) P_a \neq O(P_a \hat{d}_\mu P_a, P_a p_{\mu a} P_a).$$

To obtain a conventional Dicke Hamiltonian the two-level approximation of $O$ is taken as the right-hand-side of inequality (26) rather than the left-hand-side. This is the convention we have adopted in the form of the replacements in Eqs (17), (18) and (19). For example, if $O = H$ then the two-level model Hamiltonian in Eq. (22) is given by the right-hand-side of inequality (26).

A second (related) source of non-uniqueness in the implementation of the two-level truncation is the availability of different definitions of the material bare energy $H_m^{\alpha}$, whose eigenstates define the two-level truncation. In writing Eq. (14) we have adopted the convention of including the polarisation self-energy term $rac{e^2}{2\mu} \sum_{\mu=1}^{N} (\epsilon_{\mu,0})^2$ within the interaction Hamiltonian. Since this term depends on both $\nu$ and $N$ its inclusion in $H_m^{\alpha}$ would not provide material eigenenergies and eigenstates for which the resulting two-level model possesses a straightforwardly attainable thermodynamic limit. The convention we have adopted yields an arbitrary gauge Dicke Hamiltonian, which in commonly chosen gauges reduces to models encountered in the literature, and whose thermodynamic limit can subsequently be taken using conventional methods, such as the Holstein-Primakoff mapping.

We note that the gauge-invariance of the theory is broken by all forms of two-level truncation, regardless of what specific conventions are adopted. In Supplementary Note 6 we provide additional details regarding other conventions for the two-level truncation and show that the qualitative conclusions drawn in this work are independent of the chosen conventions.
IV. THERMODYNAMIC LIMIT OF THE ARBITRARY-GAUZE DICE MODEL

A. Hamiltonian in the normal phase

Proceeding with the above definition of the two-level truncation we now seek to determine the critical coupling point at which a phase transition occurs within the α-gauge Dicke model of Eq. (22). To this end we use a Holstein-Primakoff representation of $su(2)$ in terms of bosonic operators [5–7];

$$J_+^\alpha = b_\alpha^\dagger b_\alpha - \frac{N}{2}$$

$$J_-^\alpha = b_\alpha^\dagger \sqrt{N - b_\alpha^\dagger b_\alpha}, \quad J_\alpha^\pm = (J_\alpha^+)\dagger$$

where $[b_\alpha, b_\alpha^\dagger] = 1$. Substituting these expressions into Eq. (22) and dropping terms which vanish in the thermodynamic limit yields the Hamiltonian

$$H_{\alpha}^{\alpha,2} = \omega_m b_\alpha^\dagger b_\alpha + N\epsilon_0 + \frac{1}{2}\rho d^2 + \omega_\alpha \left( c_\alpha^\dagger c_\alpha + \frac{1}{2} \right) - C_\alpha \left( b_\alpha^\dagger + b_\alpha \right)^2 - ig_\alpha \left( b_\alpha^\dagger - b_\alpha \right) \left( c_\alpha^\dagger + c_\alpha \right) + ig_\alpha \left( b_\alpha^\dagger + b_\alpha \right) \left( c_\alpha^\dagger - c_\alpha \right).$$

As will be seen from what follows this Hamiltonian describes the so-called normal phase of the thermodynamic limit of the Dicke model.

Let us first consider the material part of $H_{\alpha}^{\alpha,2}$ on its own. This is the part that is independent of cavity canonical operators;

$$H_{\alpha}^{\alpha,2}_{\text{th},m} = \omega_m b_\alpha^\dagger b_\alpha - C_\alpha \left[ b_\alpha^\dagger + b_\alpha \right]^2 = \tilde{\omega}_m^\alpha f_\alpha^\dagger f_\alpha + \frac{1}{2}(\tilde{\omega}_m^\alpha - \omega_m)$$

where $[f_\alpha, f_\alpha^\dagger] = 1$ and

$$\tilde{\omega}_m^\alpha = \omega_m(\omega_m - 4C_\alpha).$$

The mode operators $f_\alpha, f_\alpha^\dagger$ are related to $b_\alpha$ and $b_\alpha^\dagger$ by a local Bogoliubov transformation. This has the effect of absorbing the contribution coming from $V_{\text{dip}}$ and thereby renormalising the material frequency to that in Eq. (30).

In order that the renormalised frequency $\tilde{\omega}_m^\alpha$ is real one requires that

$$\omega_m \geq 4C_\alpha = 2\rho d^2(1 - \alpha^2).$$

(31)

When the electrostatic interaction strength $C_\alpha$ is large enough this inequality may be violated, which signals a phase transition and the breakdown of the Hamiltonian describing the normal phase. We refer to this phase transition as ferroelectric, because it is completely independent of the radiative mode. It was identified within the Coulomb gauge by Keeling in Ref. [36] using a different method. Inequality (31) reduces to the result of Keeling if $\alpha = 0$, and thereby gives a generalisation to the arbitrary gauge $\alpha$. Violation of inequality (31) cannot occur in the multipolar gauge $\alpha = 1$, which does not therefore admit a purely ferroelectric phase. In what follows this finding will be reconciled with our claim that a unique phase transition is predicted within all gauges.

We now consider the thermodynamic-limit of the full Hamiltonian, $H_{\alpha}^{\alpha,2}$, which can be written in the so-called normal phase as (see Supplementary Note 3)

$$H_{\alpha}^{\alpha,2,n} = E_{\alpha}^n + f_\alpha^\dagger f_\alpha + E_{\alpha}^\prime c_\alpha^\dagger c_\alpha + \frac{1}{2}(E_{\alpha}^+ + E_{\alpha}^-) + C^n$$

(32)

where we use the superscript $n$ to refer to the normal phase. The polariton operators $f_\alpha^n, c_\alpha^n$ are bosonic satisfying $[f_\alpha^n, f_\alpha^m] = 1 = [c_\alpha^n, c_\alpha^m]$ with all other commutators between the $f_\alpha^n, c_\alpha^n$ and their conjugates vanishing. The constant $C^n$ is given by

$$C^n = N\epsilon_0 + \frac{1}{2}(\rho d^2 - \omega_m).$$

(33)

The polariton energies $E_{\alpha \pm}^n$ are given by

$$2E_{\alpha}^{n,2} = 8\tilde{g}_\alpha \tilde{g}_\alpha^\prime + \tilde{\omega}_m^\alpha + \tilde{\omega}_m^{\alpha,2} \pm \sqrt{\left( [\tilde{\omega}_m^\alpha - \omega_\alpha^2] \right)^2 + 16[\tilde{\omega}_m^\alpha \tilde{g}_\alpha + \omega_\alpha \tilde{g}_\alpha^\prime] \tilde{g}_\alpha^\prime}$$

(34)

where

$$\tilde{g}_\alpha = \sqrt{\frac{\omega_m}{\tilde{\omega}_m}} g_\alpha, \quad \tilde{g}_\alpha^\prime = \sqrt{\frac{\omega_m}{\tilde{\omega}_m}} g_\alpha^\prime.$$ 

(35)

The coupling strength at which the lower polariton energy $E_{\alpha}^-\!$ is no longer real signals the onset of a new phase, which we refer to as the abnormal phase, and which incurs the breakdown of the Hamiltonian $H_{\alpha}^{\alpha,2,n}$. Reality of $E_{\alpha}^-\!$ requires that

$$[\tilde{\omega}_m^\alpha \tilde{\omega}_m^\alpha] - 4\tilde{\omega}_m^\alpha \tilde{g}_\alpha \tilde{g}_\alpha^\prime + 16\tilde{g}_\alpha^2 \geq 0.$$ 

(37)

From the Thomas-Reiche-Kuhn (TRK) inequality

$$\frac{\epsilon^2}{m} \geq 2\omega_m d^2$$

(38)

it follows that

$$\omega_\alpha^2 = \omega^2 + \frac{\epsilon^2}{m}(1 - \alpha^2) \geq \omega^2 + 2\omega_m d^2(1 - \alpha^2) \geq 2\omega_m d^2(1 - \alpha^2).$$ 

(39)

Therefore, according to inequality (37) $E_{\alpha}^-$ is real if and only if

$$\omega_m \geq 2\rho d^2,$$ 

(40)

(continued on next page)
which defines the normal phase. Inequality (40) is a stronger bound than inequality (31). Thus, $\tilde{\omega}_m^a$, defined in Eq. (30) is also real when inequality (40) is satisfied. The two inequalities (31) and (40) are identical in and only in the Coulomb gauge $\alpha = 0$. In summary, the Hamiltonian in Eq. (32) is valid in and only in the normal phase specified by inequality (40).

For hydrogen-like atoms with $\epsilon_{n-1} \sim -e^2/(8\pi a_0^2 n^3)$, $n = 1, 2, \ldots$ and $\alpha \sim e a_0$ where $a_0$ is the Bohr radius, the critical density for a phase transition $\rho_c = \omega_m/(2\tilde{\omega}_m^2)$ is roughly $3/(64\pi a_0^3) \approx (4a_0)^{-3}$. The assumption of separate localised atoms within the EDA requires that the inter-atomic separation should be at least $2a_0$. Therefore, there is a relatively small window of validity, within which a conventional (Diele model) treatment can be expected to hold for describing the phase transition. The window is, however, finite, and as noted previously the present simplifying assumption of localised separate dipoles does not affect gauge-invariance, such that extended Dicke models [40] need not be considered for our purpose.

**B. Hamiltonian in the abnormal phase**

We now exhibit the Hamiltonian describing the system in the abnormal phase whereby inequality (40) is violated. This Hamiltonian is obtained as the thermodynamic limit of the Hamiltonian $H^{a,2}$ written in terms of alternative modes $f_\alpha$ and $c_\alpha$ such that

$$b_\alpha = f_\alpha - \sqrt{\beta_\alpha},$$

$$c_\alpha = c_\alpha' + i \sqrt{\gamma_\alpha}$$

where the parameters $\beta_\alpha$ and $\gamma_\alpha$ are assumed to be of order $N$ and are $\alpha$-dependent in general. The mode operators $b_\alpha$ are defined as before using the Holstein-Primakoff representation in Eq. (27). The derivation of the Hamiltonian $H^{a,2}_\text{th}$, where we use the superscript $^a$ to refer to the abnormal phase, is lengthy, and is given in Supplementary Note 4. The required values of $\beta_\alpha$ and $\gamma_\alpha$ are

$$\beta_\alpha = \beta := N \frac{1}{2} (1 - \tau),$$

$$\gamma_\alpha = \frac{Ng_\alpha^2}{\omega_m^2} (1 - \tau^2)$$

where

$$\tau := \frac{\omega_m \omega_m^a}{4(g_a^2 + \omega_m C_a)} = \frac{\omega_m^a}{4g_a^2 (2\rho d^2)} = \frac{\omega_m}{2\rho d^2}.$$ (45)

Note that $\beta_\alpha = \beta$ is $\alpha$-independent indicating that independent of $\alpha$ the “material” mode is always displaced by the same macroscopic quantity in the abnormal phase. On the other hand, $\gamma_\alpha$ is $\alpha$-dependent, so the extent to which the “radiative” mode is macroscopically displaced depends on the chosen definition of radiation. In particular, $\gamma_0 = 0$, so in the Coulomb gauge only the material mode is displaced. This already indicates that we should not expect there to be any macroscopic occupation of the radiative mode within the Coulomb gauge’s abnormal (ferroelectric) phase.

The final Hamiltonian is

$$H^{a,2}_\text{th} = E^{a}_\alpha f^\dagger_\alpha f_\alpha + E^{a}_\alpha c^\dagger_\alpha c_\alpha + \frac{1}{2} (E^{a}_+ + E^{a}_-) + C^a$$ (46)

where the operators $f_\alpha, c_\alpha$ are bosonic operators that satisfy $[f_\alpha, f^\dagger_\alpha] = 1 = [c_\alpha, c^\dagger_\alpha]$ with all other commutators between the $f_\alpha, c_\alpha$ and their conjugates vanishing. The constant $C^a$ is given by

$$C^a = N \left[ \epsilon_0 - \frac{\omega_m}{4\tau} (1 - \tau)^2 \right] - \frac{1}{2} \rho d^2.$$ (47)

The polariton energies $E^{a}_\pm$ are given by

$$2E^{a}_\pm = 8g_\alpha g'_\alpha + \omega_m^2 + \omega_m^a + \sqrt{\left( \omega_m^a - \omega_m \right)^2} + 16[\omega_m^a g'_\alpha + \omega_m g'_\alpha]$$ (47)

where

$$\omega_m^2 = \frac{\omega_m^2}{\gamma_\alpha} (1 - (1 - \alpha^2)\tau^2),$$

$$g'_\alpha = \sqrt{\frac{\omega_m^a}{\omega_m}} g'_\alpha,$$

$$g_\alpha = \sqrt{\frac{\omega_m^a}{\omega_m}} g_\alpha.$$ (50)

The material frequency $\omega_m^a$ is real provided

$$(2\rho d^2) \geq \omega_m^2 (1 - \alpha^2).$$ (52)

Subsequently, the lower polariton energy $E^{a}_-$ is real provided

$$\left[ \omega_m^a - \omega_m \right] - \left[ 4\omega_m^a \omega_m [g_a^2 + g'_\alpha] - 16g_a^2 g'_\alpha \right]$$

$$= (2\rho d^2) (\omega_m^2 - \omega_m (1 - \alpha^2))^2 \geq 0.$$ (53)

In the abnormal phase we have

$$2\rho d^2 \geq \omega_m,$$ (54)

implying that both $\omega_m^a - \omega_m (1 - \alpha^2) \geq 0$ and inequality (52) are satisfied. It follows that $E^{a}_-$ is necessarily real in the abnormal phase. Thus, the Hamiltonian in Eq. (46) is valid in, and only in, the abnormal phase.

At the critical coupling point where $2\rho d^2 = \omega_m$ the Hamiltonian reads

$$H^{a,2,2}_\text{th} = \sqrt{\alpha (2 - \alpha) \omega_m^2 + \omega_m^a} \left( f^\dagger_\alpha f_\alpha + \frac{1}{2} \right)$$

$$+ N\epsilon_0 - \frac{1}{2} \rho d^2.$$ (55)
The Hamiltonians in Eqs. (32), (46) and (55) describe the system in the normal phase, abnormal phase, and at the critical coupling point respectively. Their combination therefore comprises a description of the thermodynamic limit for all coupling strengths.

The lower polariton energies $E_{\alpha}^{b,a}$ constitute different two-level approximated results in each different gauge $\alpha$. However, every gauge’s approximate (Dicke) model predicts exactly one ground state phase transition, which occurs when $\omega_m = 2\rho d^2$. Furthermore, the ground state is unique within the exact (non-truncated) theory. Therefore, inequality (40), or equivalently inequality (54), should be interpreted as predicting a unique phase transition.

C. Discussion: physical nature of the unique phase transition

Although an apparently unique physical phase transition is predicted by the Dicke model in any gauge, the nature of the phase transition appears to be different depending on the value of $\alpha$. In the Coulomb gauge for example, inequalities (40) and (31) are identical, indicating that the phase transition is entirely ferroelectric; it results solely from electrostatic interactions and it can be derived without ever considering the radiative mode. In contrast, in the multipolar gauge inequality (31) cannot be violated, so while the unique physical phase transition is predicted in the multipolar gauge, it cannot be attributed to the multipolar material mode alone. As will be seen in Sec. V B the abnormal phase in the multipolar-gauge is superradiant.

The availability of different characterisations of the phase transition is not an artefact of the two-level truncation. Indeed, although within an exact theoretical treatment the Hamiltonian and its spectrum are unique, the classification of a unique phase transition as being either ferroelectric or superradiant, or some combination of the two, will naturally depend on the definitions of matter and radiation adopted. These definitions are different in different gauges. This is immediately apparent within the present arbitrary gauge framework wherein the $\alpha$-gauge “photon” number operator is defined by

$$a_\alpha^\dagger a_\alpha = \frac{\nu}{2\omega} \left[ \Pi_\alpha^2 + \omega^2 \Delta^2 \right] - \frac{1}{2} \quad (56)$$

where $\Pi_\alpha = -E_T - \alpha P_T$. The $\Pi_\alpha$ of different gauges are related by

$$\Pi_\alpha = R_{\alpha\alpha'} \Pi_{\alpha'} R_{\alpha'\alpha} = \Pi_{\alpha} - (\alpha - \alpha') P_T$$

$$= \Pi_{\alpha} - \frac{(\alpha - \alpha')}{v} \sum_{\mu=1}^{N} \epsilon(\epsilon \cdot \mathbf{d}_\mu). \quad (57)$$

In particular, the Coulomb and multipolar gauge canonical momenta differ by the gauge-invariant ($\alpha$-independent) transverse polarisation $P_T$, which can be read-off from Eq. (12):

$$\Pi_1 = R_{10} \Pi_0 R_{01} = \Pi_0 - P_T, \quad P_T = \Pi_0 - \Pi_1. \quad (58)$$

The physical meaning of “radiation” in the gauge $\alpha$ is determined by the physical meaning of the canonical momentum $\Pi_{\alpha}$. To determine this meaning let us return to the underlying multi-mode field theory as developed in Supplementary Note 1. The parameter $\alpha$ determines the transverse polarisation given by Eqs. (6) and (7) as $P_{\alpha} = \alpha P_T$, where $P_{T,\alpha} = \sum_{\mu=1}^{N} \mathbf{d}_{\mu} \delta_{\alpha}^{\mathbf{R}_{\mu}}(\mathbf{x} - \mathbf{R}_{\mu})$ is called the multipolar transverse polarisation. Meanwhile the total multipolar polarisation of $N$ dipoles including the longitudinal part is $\sum_{\mu=1}^{N} \mathbf{d}_{\mu} \delta(\mathbf{x} - \mathbf{R}_{\mu}) = \mathbf{P}_L + \mathbf{P}_T$. From Gauss’ law $\nabla \cdot \mathbf{E} = \rho$ and the defining property $\nabla \cdot \mathbf{P} = -\rho$ of the polarisation of a globally neutral system it follows that $\mathbf{P}_L = -\mathbf{E}_L$, which implies that for $\mathbf{x} \neq \mathbf{R}_\mu$ we have $\mathbf{P}_T = \mathbf{P}_L = \mathbf{E}_L$ and therefore $\Pi_\alpha = -E_T - \alpha \mathbf{E}_L$. This shows that $\alpha$ controls the relative extent to which the longitudinal electric degrees of freedom are included within the “radiation” subsystem at all points $\mathbf{x} \neq \mathbf{R}_\mu$.

In the Coulomb gauge, $\alpha = 0$, longitudinal electric degrees of freedom are completely absent from the radiative mode. The associated inter-dipole electrostatic interaction $V_{\text{dip}}$ therefore appears explicitly in the Hamiltonian as a purely material interaction, which gives rise to a purely material phase transition. In the multipolar gauge, $\alpha = 1$, longitudinal electric degrees of freedom away from the dipolar positions are implicit within the radiative mode. Inter-dipole electrostatic interactions $V_{\text{dip}}$ are therefore explicitly absent from the Hamiltonian. As a result, they cannot directly give rise to the phase transition, which instead results from coupling to the radiative mode.

It is made clear within the present arbitrary-gauge approach that these different characterisations are in no way paradoxical. In the Coulomb gauge the abnormal phase appears ferroelectric meaning that it results in a macroscopic average of the material polarisation $\mathbf{P}_T$, but not of $\mathbf{P}_0$. Since $\Pi_1 = \mathbf{P}_0 - \mathbf{P}_T$ it follows that $\Pi_1$ must possess a macroscopic average in the abnormal phase. That this is indeed the case will be confirmed in Sec. V. The Coulomb-gauge (ferroelectric) and multipolar-gauge (superradiant) characterisations of the same phase transition are therefore completely equivalent.

From the above discussion it is clear that the subsystem gauge-relativity of QED [45], is strongly exemplified by the phase transition phenomenon. In any gauge a characterisation of the phase transition can be made in terms of the “material” and “radiative” operators $b_\alpha$, $b_\alpha^\dagger$ and $a_\alpha$, $a_\alpha^\dagger$ respectively, but because these modes are mixed together by $R_{\alpha\alpha'}$ the relative extent to which the same unique phase transition is characterised as ferroelectric (material) or superradiant (radiative) naturally depends on which gauge’s definitions of light and matter are used to provide the characterisation.
A separate issue is that after the two-level approximation of the material dipoles the treatment is not exact and importantly, it is no longer gauge-invariant. This explains why the polariton energies $E_{\alpha \pm}$ are $\alpha$-dependent. More generally, it implies that when characterising the phase transition in terms of a unique physical observable, the predictions for the same observable will generally be different in different gauges [41–43]. In Sec. V, we provide expressions for the $\alpha'$-gauge’s two-level truncation of the $\alpha$-gauge canonical operators. This provides the means by which to characterise the phase transition in terms of arbitrary $\alpha$-gauge definitions of “light” and “matter”, when using the arbitrary $\alpha'$-gauge Dicke model to approximate the gauge-invariant non-truncated theory.

D. Discussion: relation to the “no-go theorem”

If in the Coulomb gauge electrostatic interactions had been neglected then rather than the inequality (37) with $\alpha = 0$, we would instead have obtained the inequality $\omega_m^2 - 2\omega_m p d^2 \geq 0$, which is necessarily satisfied according to Eq. (39). This result is in fact nothing but the usual “no-go theorem” for a superradiant phase transition in the Coulomb gauge Dicke-model [30]. An equivalent inequality is obtained by taking the ratio of terms in the square brackets on the top line of Eq. (37) rather than their difference. When taking this ratio the factor $\omega_m - 4C_0$, which when negative indicates a ferroelectric phase, appears in both the numerator and the denominator and therefore cancels out. The resulting ratio $4\rho^2/(\omega_m^2)$ often considered to be an important quantity in the context of the phase transition, is equal to $\omega_m p d^2/\omega_m^2$. It is therefore independent of $C_0$ and is bounded from above by 1, which is another expression of the “no-go theorem”. However, a phase transition will occur in the Coulomb gauge when the fortuitously cancelled quantity $\omega_m - 4C_0$ is negative.

In the $\alpha$-gauge both “A$^{2n}$” and electrostatic interaction terms are present but are weighted by $(1 - \alpha^2)^2$ and $1 - \alpha^2$ respectively. Without the inclusion of electrostatic interactions in the Coulomb-gauge Hamiltonian the corresponding term in the $\alpha$-gauge Hamiltonian would be $-\alpha^2 V_{\text{dip}}$ rather than $(1 - \alpha^2)V_{\text{dip}}$ as in Eq. (8). In this case the multipolar gauge Hamiltonian would possess a spurious term $-V_{\text{dip}}$, which would prevent a superradiant phase transition [19, 34, 41, 42]. However, with the correct interaction term $(1 - \alpha^2)V_{\text{dip}}$ the multipolar-gauge Dicke model does admit a phase transition. Our results show that the “no-go theorem” for a phase transition in traditional cavity QED systems, for which it was originally given, is actually physically baseless. It results from neglecting dipolar interactions of order $e^2$, while retaining the field self-energy “A$^{2n}$”-term, which is also of order $e^2$. If both terms are present as in the Coulomb gauge, or if both terms are absent as in the multipolar gauge, then a phase transition is predicted. More generally, in the arbitrary gauge $\alpha$ a unique phase transition is predicted when all relevant interactions are retained and all approximations are implemented self-consistently.

V. COMPARISON OF PREDICTIONS FROM DICKE MODELS OF DIFFERENT GAUGES

A. Example: the double-well dipolar potential

We now illustrate the results obtained so far by way of a concrete example. We assume that the dipole canonical operators point along the cavity polarisation $\varepsilon$ making the problem one-dimensional with $p_\alpha = \varepsilon \cdot p_\alpha$ and $r = \varepsilon \cdot r$. We assume the following double-well potential for each dipole [41]

$$V = V(\theta, \phi) = -\frac{\theta}{2} r^2 + \frac{\phi}{4} r^4$$

where $\theta$ and $\phi$ determine the shape of the double-well. The single-dipole Hamiltonian is therefore given by

$$H^\alpha_m = \frac{\mathcal{E}}{2} \left( -\partial^2_\phi - \beta \zeta^2 + \rho^2 \right)$$

where we have introduced the dimensionless variable $\zeta = r/r_0$ with $r_0 = (1/[m\phi])^{1/6}$, along with $\mathcal{E} = 1/(m r_0^2)$ and $\beta = \theta m r_0^2$. We also define the gauge-invariant dimensionless coupling parameter $\eta = \langle \varepsilon/\omega \rangle \sqrt{\rho/m}$. The parameters $\varepsilon$, $m$, and $\rho$ can now be eliminated in favour of $\mathcal{E}$, $\beta$, and $\eta$. All quantities of interest can be considered as a function of both the gauge parameter $\alpha$ and as a function of the coupling strength $\eta$ for specific choices of $\beta$ and $\mathcal{E}$. The upper and lower polariton energies are plotted in Fig. 1 as a function of $\eta$ for three fixed values of $\alpha$.

B. Average radiative quadratures and material polarisation

A clear demonstration of the gauge-relativity inherent to the quantum definitions of light and matter can be found by considering excitations of these subsystems in the thermodynamic limit of the Dicke model. We will first consider the cavity canonical operators $A = \varepsilon \cdot A$ and $\Pi_\alpha = \varepsilon \cdot \Pi_\alpha$. The transverse vector potential $A$ is manifestly gauge-invariant. In the normal phase of the thermodynamic limit $A$ vanishes. Moreover, the displacement in Eq. (42) does not alter $A$, whose thermodynamic limit therefore also vanishes in the abnormal phase. In contrast to $A$, the momentum $\Pi_\alpha$ is gauge-relative in that it represents a different physical observable for each different value of $\alpha$. In the following discussion the parameter $\alpha$ determines the degrees of freedom that are being used to define the material and radiative subsystems, i.e., the relative extent to which the transverse polarisation $P_\tau$ contributes to the radiative subsystem or
This expression happens to be $\alpha'$-independent, and is only real in the abnormal phase, which is defined by the condition $2pd^2 \geq \omega_m$.

The factor of $\alpha$ in Eq. (62) is highly significant. It follows immediately that the Coulomb-gauge canonical momentum $\Pi_0 = -\varepsilon \cdot \mathbf{E}_T$ has vanishing value in the abnormal phase independent of the gauge $\alpha'$ within which the two-level truncation is made. This confirms that with respect to the Coulomb-gauge definition of radiation the phase transition is not superradiant. On the other hand, the multipolar gauge canonical momentum $\Pi_1 = -\varepsilon \cdot (\mathbf{E}_T + \mathbf{P}_T)$ clearly possesses a non-vanishing macroscopic value in the abnormal phase for any choice of $\alpha'$, confirming that with respect to the multipolar gauge’s definition of radiation the phase transition is superradiant.

It is instructive to also calculate the $\alpha$-gauge transverse polarisation $P_{\alpha T} = \alpha \varepsilon \cdot \mathbf{P}_T = \alpha (\Pi_0 - \Pi_1)$ in the abnormal phase of the $\alpha'$-gauge Dicke model. Applying the two-level truncation given by Eq. (18) within the $\alpha'$-gauge yields

$$P_{\alpha T}^{\alpha',2} = \frac{d}{v} (J_+^{\alpha'} + J_{-}^{\alpha'}) ,$$

which also vanishes in the Coulomb gauge $\alpha = 0$. In the normal phase of the thermodynamic limit $P_{\alpha T}^{\alpha'}$ vanishes, whereas in the abnormal phase of the thermodynamic limit it reads

$$P_{\alpha T,\text{th}}^{\alpha',2} = -\frac{2\alpha d}{v} \sqrt{(N - \beta)} \beta = -\alpha pd\sqrt{1 - \tau^2}$$

where $\beta$ is defined in Eq. (43). Eqs. (62) and (64) yield

$$\Pi_{\alpha,\text{th}}^{\alpha',2} = -P_{\alpha T,\text{th}}^{\alpha',2} ,$$

which verifies the fundamental kinematic relation of the arbitrary gauge Hamiltonian theory already given in Eq. (5), namely, $\Pi_\alpha = \Pi_0 - \Pi_{\alpha T} = -\mathbf{E}_T - \mathbf{P}_{\alpha T}$. More generally, we have $\Pi_\alpha = R_{\alpha\alpha'}\Pi_{\alpha'} = \Pi_{\alpha'} - \Pi_{\alpha T} + \mathbf{P}_{\alpha T}$, from which it follows that $\Pi_{\alpha,\text{th}}^{\alpha',2} = -\Pi_{\alpha,\text{th}}^{\alpha',2} = P_{\alpha T,\text{th}}^{\alpha',2}$. This equality can also be deduced directly from Eqs. (61) and (63), and is clearly verified by the stronger equality (65).

Defining $P_T = \varepsilon \cdot \mathbf{P}_T$, Eq. (64) shows that $P_{\alpha T,\text{th}}^{\alpha',2} = P_{\alpha T,\text{th}}^{\alpha',2}/\alpha$ quantifies the distance from the critical coupling point $\tau = 1$ at which the phase transition occurs. Independent of the gauge choice $\alpha$ the onset of the abnormal phase manifests in the form of a macroscopic value

$$P_{\alpha T,\text{th}}^{\alpha',2} = -pd\sqrt{1 - \tau^2}$$

of the gauge-invariant transverse polarisation $\mathbf{P}_T$. In general, different gauges $\alpha'$ will yield different two-level model approximations of $\mathbf{P}_T$, but the thermodynamic limit $P_{\alpha T,\text{th}}^{\alpha',2}$ turns out to be $\alpha'$-independent. Within the present simplified Dicke-type treatment the field $\mathbf{P}_T$ is
The quantity $d\Pi_{\alpha,th}^{\prime,2}$ is plotted as a function of $\eta$ for $\alpha = \alpha_{JC}, 1$. As in Fig. 1, we have chosen $\beta = 2.4$, and then chosen $\xi$ such that $\omega_m = \omega$. The definition of the canonical momentum changes linearly with $\alpha$ from $\alpha = 0$ such that $\Pi_0 = -E_T$ to $\alpha = 1$ such that $\Pi_1 = -D_T$. Correspondingly, for fixed $\alpha$ we have $\Pi_{\alpha,th}^{\prime,2}/\Pi_{\alpha,th}^{\prime,2} = \alpha/\alpha'$. Thus, the ratio of the magnitudes of the two curves is constant with value $\alpha_{JC}/1 = \alpha_{JC}$. Note in addition, that $\Pi_{\alpha,th}^{\prime,2} = P_{\alpha,th}^{\prime,2}$, meaning that the curve corresponding to $\alpha = 1$ illustrates the gauge-invariant manifestation of the abnormal phase via the transverse polarisation $P_T$.

independent of spatial position $x$, but at a more fundamental level Eq. (7) shows that $P_T$ coincides with the longitudinal electric field $E_L$ away from the dipole positions, i.e., for $x \neq R_\mu$. Whether one considers $E_L = P_T$ to be “material” or “radiative” determines whether one calls the phase transition “purely ferroelectric” or “superradiant”, as discussed in Sec. IV C.

The extension of the Dicke model to arbitrary gauges is non-trivial, not only because it allows us to prove general results holding in any gauge, but also because there exist gauges of physical significance such that $0 < \alpha < 1$. For example, the Jaynes-Cummings gauge $\alpha_{JC}$ identified in [43] (see also [46–48]) is noteworthy, because it eliminates counter-rotating terms from the linear matter-radiation interaction Hamiltonian. Even in the ultrastrong-coupling regime, ground state photon population is highly suppressed in this gauge. Moreover, for the simple case of a single dipole it was shown that there exist regimes for which the Jaynes-Cummings-gauge two-level model offers a significantly better representation of the Hamiltonian ground state than the Coulomb and multipolar gauge quantum Rabi models (see also Supplementary Note 7).

Considering the example of a double-well potential for each dipole as in Sec. V A, Fig. 2 plots $d\Pi_{\alpha,th}^{\prime,2}$ as a function of $\eta$ for $\alpha = \alpha_{JC}$ and $\alpha = 1$. At resonance, the ratio $\Pi_{\alpha_{JC},th}^{\prime,2}/\Pi_{\alpha,th}^{\prime,2} = \alpha_{JC}$ is 1/2 if $\eta = 0$. The value of $\alpha_{JC}$ decreases as $\eta$ increases, but remains roughly 1/2 over the range of $\eta$ in Fig. 2. Thus, in the Jaynes-Cummings gauge the radiative macroscopic manifestation of the phase transition is roughly halfway between that of the Coulomb and multipolar gauges. It is known that in the rotating-wave approximated multipolar-gauge theory the phase transition location is shifted by a factor of 1/2. However, if counter-rotating terms are instead eliminated through the choice of gauge then there is a non-vanishing electrostatic component within the Hamiltonian. This interaction evidently performs the same function as the multipolar-gauge counter-rotating terms in yielding a phase transition at the same location $\omega_m = 2\rho d^2$.

VI. NUMERICAL RESULTS FOR FINITE $N$

Here we provide numerical results for selected low energy properties of the system for various values of $N$. The first excited and ground state eigenenergies of the total Hamiltonian are denoted $E$ and $G$ respectively. We again consider the double-well potential example. We consider the thermodynamic limit of the first transition
energy $E - G$, and the normalised second derivative of the ground energy $N^{-1}d^2G_s/d\eta^2$ where $G_s = G - \rho d^2/2$. The coupling-dependent shift $\rho d^2/2$ in Eq. (22) shifts $d^2G/d\eta^2$ by the constant amount $m\omega^2d^2/\epsilon^2$. It is therefore more convenient to consider the second derivative of $G_s/N$ rather than that of $G/N$ when comparing finite $N$ results with the thermodynamic limit. According to Eqs. (33) and (47) the thermodynamic limit of the $\alpha$-gauge Dicke model gives $N^{-1}d^2G^{\alpha,2}_s/d\eta^2|_{\text{th},n=0}$ in the normal phase, and
\begin{equation}
\frac{1}{N} d^2G^{\alpha,2}_s \bigg|_{\text{th},n=0} = -\omega_m \frac{d^2}{d\eta^2} \frac{(1 - \tau)^2}{4\tau} \tag{67}
\end{equation}
in the abnormal phase. Being second order, the phase transition is located by a discontinuity in the thermodynamic limit of $N^{-1}d^2G^{\alpha,2}_s/d\eta^2$ [7].

We choose $\beta = 3.3$, which provides a highly anharmonic single-dipole spectrum such that $(\epsilon_2 - \epsilon_0)/\omega_m \approx 36$. The two-level truncation within the multipolar gauge $\alpha = 1$ is subsequently found to be accurate in predicting low energy properties. This was already confirmed in the case of the Rabi model $N = 1$ in Ref. [41]. For the same double-well potential example, the multipolar gauge two-level truncation actually becomes more accurate as $N$ increases. For values of $N$ up to $N = 4$, Fig. 3a compares the multipolar-gauge two-level model predictions of the transition energy $E - G$ with corresponding predictions of the non-truncated theory. The latter are obtained numerically by retaining a number of higher material levels. From these results it is reasonable to assume that the thermodynamic limit of the multipolar gauge Dicke model is also accurate. For finite $N$, the variation of $N^{-1}d^2G_s/d\eta^2$ constitutes a precursor to the discontinuity that locates the phase transition in the thermodynamic limit. Curves corresponding to different $N$ intersect at the phase transition point, as shown in Fig. 3b.

The double-well might also be parameterised differently. For example, choosing $\beta = 1.5$ reduces the anharmonicity significantly, such that $(\epsilon_2 - \epsilon_0)/\omega_m \approx 3.2$. In this case, single-dipole two-level models are only able to remain accurate in predicting low energy properties. The multipolar gauge is typically sub-optimal for this purpose whereas the Jaynes-Cummings gauge is accurate [43]. Illustrative results for the case $\beta = 1.5$ and $N = 1$ are given in Supplementary Note 7. For $N > 1$ the situation changes such that two-level models become largely inaccurate (see Supplementary Note 7).

The situation may also change if additional cavity modes are taken into account [49, 50]. In particular, the multipolar-gauge coupling scales as $\sqrt{\omega}$ such that the single-mode approximation appears least favourable in this gauge, and has been shown to breakdown in the ultrastrong-coupling regime [49]. To incorporate some of the effects of non-resonant modes while still remaining within a single-mode description a formal procedure of adiabatic elimination of the non-resonant modes could be used. This also has the advantage of enabling an exploration of more diverse dipolar geometries [29]. However, such a procedure cannot straightforwardly be performed in such a way as to preserve gauge-invariance.

in this work we have been concerned with unambiguously determining whether a physical phase transition occurs, and on subsequently understanding how its cause and physical nature are related to the choice of gauge. For this purpose a single-mode restriction that preserves gauge-invariance is obviously necessary, but is also sufficient, because the qualitative results of the thermodynamic limit of the single-mode Dicke model are known to carry over to the multi-mode case [51]. A detailed analysis for the determination of optimal multi-mode Dicke models in the thermodynamic limit, including the interplay between the choice of gauge, the single-mode restriction, and the two-level truncation, will be given elsewhere.

\section{VII. Conclusions}

We have shown that a unique physical phase transition will occur in simple many-dipole cavity QED systems, and we have given a coherent picture of this phase transition using an arbitrary-gauge formulation of QED. We have resolved all ambiguities pertaining to the choice of gauge by determining both the origin and properties of the phase transition in terms of any gauge’s definitions of the quantum subsystems, and by demonstrating consistency between all gauge choices. In so doing we have clearly shown that the “no-go theorem” does not apply, and that specific artificial systems are not required to support a phase transition. We have shown that although the two-level approximation ruins the gauge-invariance of the theory, unambiguous predictions can be obtained. For a specific example, we have exhibited accurate approximate predictions for finite numbers of dipoles, with analogous predictions in the thermodynamic limit.

The phase transition results from a correct treatment of electrostatic interactions, and manifests unambiguously via a macroscopic average of the gauge-invariant transverse polarisation field $P_T$. The extent to which the phase transition is classed as purely ferroelectric or super-radiant depends on the extent to which $P_T$ contributes to the radiative subsystem. The Coulomb gauge description of QED includes the longitudinal electric degrees of freedom within the material quantum subsystem, and as a result the phase transition appears purely ferroelectric. The multipolar gauge description of QED includes the same degrees of freedom implicitly within the radiative subsystem such that the phase transition appears super-radiant. The more general $\alpha$-gauge constitutes a mixture of these two extremes, and supports notable alternatives to the usual gauges specified by values $0 < \alpha < 1$.

The framework developed here should be straightforwardly extendable to the description of artificial solid-state and superconducting systems, as well as to driven and dissipative systems. It is also of practical importance
to extend the treatment here to the multi-mode setting and to ascertain optimal approximate (Dicke) models in this case. The arbitrary-gauge approach is potentially applicable to elucidate both qualitatively and quantitatively, the underlying causes and the physical natures, of thermodynamic phase transitions in various light-matter systems, and in each case, to determine optimal approximate descriptions.
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Supplementary Information - Uniqueness of the phase transition in many-dipole systems

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Supplementary Note 1: Arbitrary gauge quantisation of the matter-radiation system

Throughout this section we will frequently use the Helmholtz decomposition of a vector field $\mathbf{V}$ into transverse and longitudinal parts $\mathbf{V}_T$ and $\mathbf{V}_L$ such that for all $x$

$$\mathbf{V} = \mathbf{V}_T + \mathbf{V}_L, \tag{68}$$

$$\nabla \cdot \mathbf{V}_T(x) = 0, \tag{69}$$

$$\nabla \times \mathbf{V}_L(x) = 0. \tag{70}$$

We assume that all vector fields vanish at the boundaries $|x| \to \infty$, which allows free use of integration by parts such as

$$\int d^3x \mathbf{V}(x) \cdot \nabla f(x) = -\int d^3x f(x) \nabla \cdot \mathbf{V}(x). \tag{71}$$

Recalling that $\nabla \times \nabla f(x) = 0$ for any $f$ and for all $x$, we have that for any longitudinal field $\mathbf{U}_L$ there exists an $f$ such that $\mathbf{U}_L = \nabla f$. It follows from Eq. (71) that

$$\int d^3x \mathbf{V}_T(x) \cdot \mathbf{U}_L(x) = 0 \tag{72}$$

for any vector fields $\mathbf{V}$ and $\mathbf{U}$. These formulae will be frequently used in what follows.

We consider $N$ hydrogen-like non-relativistic atoms comprised of positive charges $e_p = e$ with mass $m_p$ at positions $\mathbf{r}_{p\mu}$, each of which is paired with a negative charge $e_e = -e$ with mass $m_e$ at $\mathbf{r}_{e\mu}$. The four-current has components $(j^a) = (\rho, \mathbf{J})$ with

$$\rho(x) = \sum_{\mu=1}^{N} \rho_\mu(x) = \sum_{\mu=1}^{N} \sum_{\sigma=e,p} e_\sigma \delta(x - \mathbf{r}_{\sigma\mu}) = e \sum_{\mu=1}^{N} \left[ \delta(x - \mathbf{r}_{p\mu}) - \delta(x - \mathbf{r}_{e\mu}) \right], \tag{73}$$

$$\mathbf{J}(x) = \sum_{\mu=1}^{N} \mathbf{J}_\mu(x) = \sum_{\mu=1}^{N} \sum_{\sigma=e,p} e_\sigma \mathbf{r}_{\sigma\mu} \delta(x - \mathbf{r}_{\sigma\mu}) = e \sum_{\mu=1}^{N} \left[ \mathbf{r}_{p\mu} \delta(x - \mathbf{r}_{p\mu}) - \mathbf{r}_{e\mu} \delta(x - \mathbf{r}_{e\mu}) \right], \tag{74}$$

such that $\partial_a j^a = \dot{\rho}(x) + \nabla \cdot \mathbf{J} = 0$. Since the system is globally neutral we can define the polarisation field $\mathbf{P}$ by the equation $-\nabla \cdot \mathbf{P} = \rho$, which can be solved to give

$$\mathbf{P}(x) = -\int d^3x' \mathbf{g}(x, x') \rho(x') = -\sum_{\mu=1}^{N} \int d^3x' \mathbf{g}(x, x') \rho_\mu(x') \equiv \sum_{\mu=1}^{N} \mathbf{P}_\mu(x) \tag{75}$$

where

$$\nabla \cdot \mathbf{g}(x, x') = \delta(x - x') \tag{76}$$

defines the green’s function $\mathbf{g}$ for the divergence operator. Since $\nabla \cdot \mathbf{g}(x, x') = \nabla \cdot \mathbf{g}_L(x, x')$, Eq. (75) only fixes $\mathbf{g}_L = \mathbf{g} - \mathbf{g}_T$ uniquely as

$$\mathbf{g}_L(x, x') = -\nabla \frac{1}{4\pi|x - x'|}. \tag{77}$$

Any field $\mathbf{g}_T$ with $\nabla \cdot \mathbf{g}_T(x, x') = 0$, can be added to $\mathbf{g}_L$ in Eq. (77) to obtain a $\mathbf{g}$ that satisfies Eq. (75). It follows that $\mathbf{P}_L$ is fixed uniquely by Eqs. (75) and (77) while $\mathbf{P}_T$ is arbitrary, being determined by Eq. (75) and $\mathbf{g}_T$. To avoid any confusion we note that here we are using the notation $\mathbf{P}_T$ to refer to the arbitrary transverse part of $\mathbf{P}$ given in Eq. (75). In the main text we reserve the notation $\mathbf{P}_T$ for specifically the multipolar transverse polarisation, which is just one possible example of the transverse part of $\mathbf{P}$.

Assuming for generality an external potential $V_{\text{ext}}$ acting on the charges the standard Lagrangian describing the system of all charges coupled to the Maxwell field is given by

$$L = \frac{1}{2} \sum_{\mu=1}^{N} \left[ m_p \dot{\mathbf{r}}_{p\mu}^2 + m_e \dot{\mathbf{r}}_{e\mu}^2 \right] - V_{\text{ext}} - \int d^3x \left[ j^a(x) A_a(x) + \frac{1}{4} F_{ab}(x) F^{ab}(x) \right]. \tag{78}$$
where \((A^a) = (A_0, A)\) are the components of the electromagnetic four-potential and \(F_{ab} = \partial_a A_b - \partial_b A_a\). The field tensor components \(F_{ab}\) are invariant under a gauge transformation \(A_0 \rightarrow A_0 - \partial_a \chi\) where \(\chi\) is an arbitrary function. We encode this gauge-freedom into the arbitrary function \(g\) by defining

\[
\chi(x) = \int d^3x' g(x', x) \cdot A_T(x') \equiv \int d^3x' g_T(x', x) \cdot A_T(x') \tag{79}
\]

and by subsequently defining the arbitrary potentials

\[
A_0 = \phi_{\text{Coul}} - \partial_t \chi, \tag{80}
\]

\[
A = A_T + A_L \equiv A_T + \nabla \chi \tag{81}
\]

where \(A_T\) is the gauge-invariant transverse vector potential and

\[
\phi_{\text{Coul}}(x) = \int d^3x' \frac{\rho(x')}{4\pi|x - x'|}. \tag{82}
\]

The choice \(g_T = 0\) defines the Coulomb gauge wherein \(A = A_T\) and \(A_0 = \phi_{\text{Coul}}\). A different gauge can be specified by choosing a different \(g_T\).

The Lagrangian \(L\) in Eq. (78) is not gauge-invariant. We therefore define the equivalent, but gauge-invariant Lagrangian

\[
L' = L - \frac{d}{dt} \int d^3x P(x) \cdot A(x). \tag{83}
\]

Using Eqs. (80) and (81) in conjunction with Eq. (79) the Lagrangian \(L'\) can be written

\[
L' = L_0 - \frac{d}{dt} \int d^3x P_T(x) \cdot A_T(x) \tag{84}
\]

where

\[
L_0 = \frac{1}{2} \sum_{\mu=1}^{N} [m_p r_{p\mu}^2 + m_e r_{e\mu}^2] - V_{\text{ext}} - \frac{1}{2} \int d^3x \rho(x) \phi_{\text{Coul}}(x) + \int d^3x J(x) \cdot A_T(x) + \frac{1}{2} \int d^3x [E_T(x)^2 - B(x)^2] \tag{85}
\]

in which \(E_T = -\dot{A}_T\) and \(B = \nabla \times A_T\). The remaining total time derivative in Eq. (84) depends on \(P_T\) which according to Eq. (75) is uniquely determined through a choice of gauge \(g_T\). It is straightforward to show that

\[
\int d^3x P_T(x) \cdot A_T(x) = -\int d^3x \rho(x) \chi(x), \tag{86}
\]

where now according to Eq. (79) it is the arbitrary function \(\chi\) that is determined by the gauge \(g_T\). Note that \(P_T\) in Eq. (84) is a completely arbitrary transverse field and need not coincide with the usual multipolar transverse polarisation field. In writing Eq. (85) we have used the Gauss law \(\nabla \cdot E_L = -\nabla^2 \phi_{\text{Coul}} = \rho\) and integration by parts to separate the transverse and electrostatic parts of the electromagnetic Lagrangian as

\[
-\frac{1}{4} \int d^3x F_{ab} F^{ab} = \frac{1}{2} \int d^3x [E(x)^2 - B(x)^2] = \frac{1}{2} \int d^3x [E_T(x)^2 - B(x)^2] + \frac{1}{2} \int d^3x \rho(x) \phi_{\text{Coul}}(x). \tag{87}
\]

Combining the electrostatic part of Eq (87) with the term \(\int d^3x \rho\phi_{\text{Coul}}\) coming from the component \(-\int d^3x A_0 \rho\) of the interaction Lagrangian we obtain the final electrostatic interaction term \(-\int d^3x \rho \phi_{\text{Coul}}/2\) that appears in Eq. (85).

We now introduce relative and centre-of-mass coordinates for each atom \(\mu\), which are defined by

\[
\mathbf{r}_\mu = \mathbf{r}_{\mu_0} - \mathbf{r}_{\mu_0} \tag{88}
\]

\[
\mathbf{R}_\mu = \frac{m_e \mathbf{r}_{\mu_0} + m_p \mathbf{r}_{\mu_0}}{m_e + m_p} \tag{89}
\]

along with total and reduced masses defined by

\[
M = m_e + m_p, \tag{90}
\]

\[
m = \frac{m_e m_p}{M}. \tag{91}
\]
Similarly to Eq. (95) the multipole expansion of the current yields to leading order
\[
\frac{1}{2} \int d^3x \rho(x) \phi_{\text{Coal}}(x) = \int d^3x \int d^3x' \frac{\rho(x)\rho(x')}{8\pi|x-x'|} = \sum_{\mu=1}^{N} V_{\mu} + \sum_{\mu \neq \nu}^{N} \int d^3x \int d^3x' \frac{\rho_{\mu}(x)\rho_{\nu}(x')}{8\pi|x-x'|}
\]  
(92)
where
\[
V_{\mu} = V_{\mu}^{\text{self}} - \sum_{\mu = 1}^{N} \frac{e^2}{4\pi|\mathbf{r}_{\mu}|}.
\]
(93)
The first term on the right-hand-side in Eq. (93) gives the divergent Coulomb self-energies of the charges at \( r_{\sigma \mu} \) and \( r_{p\mu} \). The second term gives the potential energy that binds the charge \(-e\) at \( r_{\sigma \mu} \) to its nucleus \(+e\) at \( r_{p\mu} \). The second term on the right-hand-side of Eq. (92) gives the inter-atomic Coulomb interactions.

We now restrict our attention to gauges in which the arbitrary transverse polarisation takes the form of a weighted multipolar transverse polarisation with weight \( \alpha \);
\[
P_{T,i}(x) = \sum_{\mu=1}^{N} P_{T,\mu,i}(x) = \alpha \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma}(r_{\sigma \mu} - R_{\mu}) \int_{0}^{1} d\lambda \delta_{ij}^{T}(x - R_{\mu} - \lambda[r_{\sigma \mu} - R_{\mu}]) =: P_{T,\alpha,i}(x)
\]
(94)
where \( \alpha \) is real and dimensionless and where \( P_{T,\mu} \) denotes the transverse polarisation associated with the \( \mu \)th atom expressed in terms of the centre-of-mass position \( R_{\mu} \). With this restriction the gauge is now completely determined by selecting a value of \( \alpha \). The derivation of the Dicke-model also requires the electric-dipole approximation (EDA), which for simplicity we implement at this stage rather than later on. In the EDA the charge density within the inter-atomic Coulomb interaction is approximated by the first non-zero term in the multipole expansion of the single atom density \( \rho_{\mu} \) about the atomic centre-of-mass at \( R_{\mu}, \) viz.,
\[
\rho(x) = \sum_{\mu=1}^{N} \rho_{\mu}(x) = \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma}\delta(x - R_{\mu} - (r_{\sigma \mu} - R_{\mu}))
\]
\[
= \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma} [\delta(x - R_{\mu}) - (r_{\sigma \mu} - R_{\mu}) \cdot \nabla \delta(x - R_{\mu}) + ...] \approx -\sum_{\mu=1}^{N} \mathbf{d}_{\mu} \cdot \nabla \delta(x - R_{\mu})
\]
(95)
where \( \mathbf{d}_{\mu} = -e\mathbf{r}_{\mu} \) is the dipole moment of the \( \mu \)th dipole. We thereby obtain
\[
\sum_{\mu \neq \nu}^{N} \int d^3x \int d^3x' \frac{\rho_{\mu}(x)\rho_{\nu}(x')}{8\pi|x-x'|} \approx \frac{1}{2} \sum_{\mu \neq \nu}^{N} \mathbf{d}_{\mu,i}\mathbf{d}_{\nu,j}\delta_{ij}^{T}(R_{\mu} - R_{\nu}) =: V_{\text{dip}}.
\]
Similarly to Eq. (95) the multipole expansion of the current yields to leading order
\[
\mathbf{J}(x) = \sum_{\mu=1}^{N} \mathbf{J}_{\mu}(x) = \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma} \mathbf{r}_{\sigma \mu} \delta(x - R_{\mu} - (r_{\sigma \mu} - R_{\mu}))
\]
\[
= \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma} \mathbf{r}_{\sigma \mu} [\delta(x - R_{\mu}) - (r_{\sigma \mu} - R_{\mu}) \cdot \nabla \delta(x - R_{\mu}) + ...] \approx \sum_{\mu=1}^{N} \mathbf{d}_{\mu} \delta(x - R_{\mu}).
\]
(97)
The current-dependent interaction component of \( L_{0} \) in Eq. (85) therefore becomes in the EDA
\[
\int d^3x \mathbf{J}(x) \cdot \mathbf{A}_{T}(x) \approx \sum_{\mu=1}^{N} \mathbf{d}_{\mu} \cdot \mathbf{A}_{T}(R_{\mu}).
\]
(98)
The multipole expansion of the \( \alpha \)-dependent transverse polarisation field in Eq. (94) yields to leading order
\[
P_{T,\alpha,i}(x) = \alpha \sum_{\mu=1}^{N} \sum_{\sigma = e, p} \epsilon_{\sigma}(r_{\sigma \mu} - R_{\mu}) \int_{0}^{1} d\lambda [1 - \lambda(r_{\sigma \mu} - R_{\mu}) \cdot \nabla + ...] \delta_{ij}^{T}(x - R_{\mu})
\]
\[
\approx \alpha \sum_{\mu=1}^{N} \mathbf{d}_{\mu,i}\delta_{ij}^{T}(x - R_{\mu}).
\]
(99)
At this stage we neglect the nuclear motions $\dot{R}_\mu$, such that when we arrive at the quantum theory $R_\mu$ will simply denote the fixed classical position of the $\mu$'th dipole. The polarisation-dependent interaction component of $L'$ in Eq. (84) therefore becomes

$$-\frac{d}{dt} \int d^3x \mathbf{P}_{T\alpha}(x) \cdot \mathbf{A}_T(x) = -\alpha \frac{d}{dt} \sum_{\mu=1}^N d_\mu \cdot \mathbf{A}_T(R_\mu) = -\alpha \sum_{\mu=1}^N \left[ \dot{d}_\mu \cdot \mathbf{A}_T(R_\mu) + d_\mu \cdot \dot{\mathbf{A}}_T(R_\mu) \right].$$

(100)

 Altogether Eqs. (96), (98) and (99) yield the Lagrangian $L'$ within the EDA as

$$L' = \sum_{\mu=1}^N \frac{1}{2} m R_\mu^2 - V - V_{dip} + \sum_{\mu=1}^N \left[ (1 - \alpha) \dot{d}_\mu \cdot \mathbf{A}_T(R_\mu) - \alpha d_\mu \cdot \dot{\mathbf{A}}_T(R_\mu) \right] + \frac{1}{2} \int d^3x \left[ \mathbf{E}_T(x)^2 - \mathbf{B}(x)^2 \right] =: L_\alpha$$

(101)

where we have absorbed the external potential $V_{ext}$ into the definition of the total intra-atomic potential as

$$V := \sum_{\mu=1}^N V_\mu + V_{ext}.$$  

(102)

The electrostatic energies $V_\mu$ and $V_{dip}$ are defined in Eqs. (93) and (96) respectively, while the transverse electric and magnetic fields are given by $\mathbf{E}_T = -\dot{\mathbf{A}}_T$ and $\mathbf{B} = \nabla \times \mathbf{A}_T$ respectively. Thus, the Lagrangian in Eq. (101) is fully specified in terms of the dynamical variable set $\{r_\mu, \dot{r}_\mu, \mathbf{A}_T, \dot{\mathbf{A}}_T\}$ together with the fixed dipolar positions $R_\mu$.

It is now possible to switch to the canonical formalism by defining the canonical momenta

$$p_{\mu\alpha} = \frac{\partial L_\alpha}{\partial \dot{r}_\mu},$$

(103)

$$\Pi_{T\alpha} = \frac{\delta L_\alpha}{\delta \dot{\mathbf{A}}_T}$$

(104)

and to then quantise the theory by assuming the canonical commutation relations

$$[r_{\mu,i}, p_{\nu,j}] = i \delta_{\mu\nu} \delta_{ij},$$

(105)

$$[A_{T,i}(x), \Pi_{T,j}(x')] = i \delta_{ij} \delta(x - x').$$

(106)

The centre-of-mass variable $R_\mu$ is a classical position of the $\mu$'th dipole. The $\alpha$-dependent canonical momenta are found to be

$$p_{\mu\alpha} = m \dot{R}_\mu - e(1 - \alpha) \mathbf{A}_T(R_\mu),$$

(107)

$$\Pi_{T\alpha} = -\mathbf{E}_T - P_{T\alpha}.$$  

(108)

For any two values $\alpha$ and $\alpha'$ of the gauge parameter the canonical operators are related by the unitary gauge-fixing transformation $R_{\alpha\alpha'}$ as

$$p_{\mu\alpha} = R_{\alpha\alpha'} p_{\mu\alpha'} R_{\alpha\alpha'}^{-1},$$

(109)

$$\Pi_{T\alpha} = R_{\alpha\alpha'} \Pi_{T\alpha'} R_{\alpha\alpha'}^{-1} \]$$

(110)

where

$$R_{\alpha\alpha'} = \exp \left[ i(\alpha - \alpha') \sum_{\mu=1}^N d_\mu \cdot \mathbf{A}_T(R_\mu) \right].$$

(111)

The Hamiltonian is defined by

$$H = \sum_{\mu=1}^N \dot{R}_\mu \cdot p_{\mu\alpha} + \int d^3x \dot{\mathbf{A}}_T(x) \cdot \Pi_{T\alpha}(x) - L_\alpha.$$  

(112)
Through substitution of Eqs. (4) and (5) into Eq. (112) the Hamiltonian written in terms of the manifestly gauge-invariant operators \{r_{\mu}, \mathbf{r}_{0}, \mathbf{A}_T, \mathbf{A}_T\} is found to coincide with the total energy expressed as the sum of material and transverse-electromagnetic energies:

\[
H = E_{\text{matter}} + E_{\text{field}} = \left[ \sum_{\mu=1}^{N} \frac{1}{2m} \mathbf{p}_{\mu}^2 + V + V_{\text{dip}} \right] + \frac{1}{2} \int d^3 x \left[ \mathbf{E}_T(x)^2 + \mathbf{B}(x)^2 \right].
\]

(113)

While this expression is clearly \(\alpha\)-independent (gauge-invariant), when expressed in terms of canonical operators the Hamiltonian has an \(\alpha\)-dependent functional form given by

\[
H = \sum_{\mu=1}^{N} \frac{1}{2m} \left[ \mathbf{p}_{\mu} + e(1-\alpha)\mathbf{A}_T(\mathbf{r}_\mu) \right]^2 + V + \alpha^2 \sum_{\mu=1}^{N} \frac{d_{\mu,i}d_{\mu,j}\delta_{ij}}{2} \delta(0) + (1-\alpha^2)V_{\text{dip}} + \alpha \sum_{\mu=1}^{N} \mathbf{d}_\mu \cdot \mathbf{\Pi}_{\alpha}(\mathbf{r}_\mu)
\]

\[
+ \frac{1}{2} \int d^3 x \left[ \mathbf{P}_{\alpha}(x)^2 + \mathbf{B}(x)^2 \right].
\]

(114)

In writing Eq. (114) we have used

\[
\frac{1}{2} \int d^3 x \mathbf{P}_{\alpha}(x)^2 = \frac{\alpha^2}{2} \sum_{\mu,\nu=1}^{N} d_{\mu,i}d_{\nu,j}\delta_{ij}(\mathbf{R}_{\mu} - \mathbf{R}_{\nu}) = \frac{\alpha^2}{2} \sum_{\mu=1}^{N} d_{\mu,i}d_{\nu,j}\delta_{ij}(0) - \alpha^2 V_{\text{dip}},
\]

(115)

which follows from Eqs. (96) and (99), together with the property \(\delta_{ij}(x) = -\delta_{ij}(x)\) for \(x \neq 0\). The Hamiltonian reduces to the Coulomb-gauge result if we choose \(\alpha = 0\). In this case direct electrostatic interactions are fully explicit in the form of the dipole-dipole interaction \((1-\alpha^2)V_{\text{dip}} = V_{\text{dip}}\). The field degrees of freedom are defined in terms of the transverse vector potential and its velocity, the transverse electric field; \(\mathbf{\Pi}_{T0} = \mathbf{A}_T = -\mathbf{E}_T\). Another common choice of gauge is the multipolar gauge obtained by choosing \(\alpha = 1\). In this gauge electrostatic interactions are eliminated; \((1-\alpha^2)V_{\text{dip}} = 0\), while the field degrees of freedom are defined in terms of the transverse vector potential and the retarded transverse displacement field; \(\mathbf{\Pi}_{T1} = -\mathbf{D}_T = -\mathbf{E}_T - \mathbf{P}_{T1}\). Outside of the atoms the transverse displacement field coincides with the total electric field, which in the EDA means that \(\mathbf{D}_T(x) = \mathbf{E}(x)\) for \(x \neq \mathbf{R}_\mu\). More generally, in the \(\alpha\)-gauge the Hamiltonian has a hybrid form. Coulomb-gauge matter-transverse field interaction terms are weighted by \((1-\alpha)\) while multipolar-gauge matter-transverse field interaction terms are weighted by \(\alpha\). Electrostatic interaction terms are weighted by \(1-\alpha^2\). In the case \(N = 2\) the Hamiltonian in Eq. (114) coincides with that given in Ref. [48].

The above expressions are applicable for general field operators \(\mathbf{A}_T\) and \(\mathbf{\Pi}_{T\alpha}\). We now define the operator

\[
a_{\alpha,\lambda}(k) := \sqrt{\frac{1}{2\omega}} \left( \omega \tilde{\mathbf{A}}_{\alpha,\lambda}(k) + i\mathbf{\Pi}_{\alpha,\lambda}(k) \right)
\]

(116)

where \(\tilde{\mathbf{A}}_{\alpha,\lambda}(k) = \epsilon_{\lambda}(k) \cdot \tilde{\mathbf{A}}(k)\) and \(\tilde{\mathbf{\Pi}}_{\alpha,\lambda}(k) = \epsilon_{\lambda}(k) \cdot \tilde{\mathbf{\Pi}}(k)\). Here tildes denote the Fourier transform and \(\epsilon_{\lambda}(k)\), \(\lambda = 1, 2\) are mutually orthogonal unit vectors both orthogonal to \(k\). From the transverse canonical commutation relation

\[
[A_{\alpha,i}(x), \Pi_{\alpha,j}(x')] = i\delta_{ij}(x - x')
\]

(117)

it follows that

\[
[a_{\alpha,\lambda}(k), a_{\alpha,\lambda}^\dagger(k')] = \delta_{\lambda\lambda'}\delta(k - k').
\]

(118)

The operators \(a_{\alpha,\lambda}(k)\) and \(a_{\alpha,\lambda}^\dagger(k)\) are recognisable as annihilation and creation operators for a photon with momentum \(k\) and polarisation \(\lambda\). In terms of these operators the canonical fields support the Fourier representations

\[
\mathbf{A}_T(x) = \int d^3 k \sum_{\lambda} g_{\lambda}(k) \left( a_{\alpha,\lambda}^\dagger(k)e^{-ik\cdot x} + a_{\alpha,\lambda}(k)e^{ik\cdot x} \right),
\]

\[
\mathbf{\Pi}_{T\alpha}(x) = i \int d^3 k \sum_{\lambda} \omega_{\lambda}(k) \left( a_{\alpha,\lambda}^\dagger(k)e^{-ik\cdot x} - a_{\alpha,\lambda}(k)e^{ik\cdot x} \right)
\]

(119)

where \(\omega = |k|\) and \(g := 1/\sqrt{2\omega(2\pi)^3}\).
If we assume an implicit cavity with volume \( v \) that satisfies periodic boundary conditions, the continuous label \( k \) becomes discrete. The pair \( k\lambda \) then labels a radiation mode and the operators \( a_{\alpha,\lambda}(k) \) are labelled with discrete index \( \alpha \), as \( a_{\alpha,k\lambda} \), which satisfy

\[
[a_{\alpha,k\lambda}, a_{\beta,k'\lambda}^\dagger] = \delta_{\lambda\lambda'}\delta_{kk'}. \tag{120}
\]

As a less realistic, but simpler model for the cavity we may restrict our attention to a single fixed mode \( k\lambda \) and ignore all modes \( k'\lambda' \neq k\lambda \). In this case the field operators become

\[
A_T(x) = g\varepsilon \left( a_{\alpha}^\dagger e^{-ik\cdot x} + a_{\alpha} e^{ik\cdot x} \right),
\]

\[
\Pi_{T\alpha}(x) = i\omega g\varepsilon \left( a_{\alpha}^\dagger e^{-ik\cdot x} - a_{\alpha} e^{ik\cdot x} \right),
\]

where \( g = 1/\sqrt{2\omega v}, \) \( \omega = |k|, \) \( \varepsilon \equiv \varepsilon_k\lambda, \) and \( a_{\alpha} \equiv a_{\alpha,k\lambda} \) with \( [a_{\alpha}, a_{\beta}^\dagger] = 1 \). Eqs. (121) and (122) imply that the cavity canonical operators now satisfy the commutation relation

\[
[A_{T,i}(x), \Pi_{T\alpha,j}(x')] = \frac{i\varepsilon_i\varepsilon_j}{v} \cos [k \cdot (x - x')]. \tag{123}
\]

To preserve Eq. (117) we must discretise the \( k \)-space representation of the transverse delta-function and subsequently perform the single-mode approximation as

\[
\delta_T^{ij}(x) = \int \frac{d^3k}{(2\pi)^3} \sum_{\lambda=1,2} \varepsilon_{\lambda,i}(k)\varepsilon_{\lambda,j}(k) \cos(k \cdot x) \rightarrow \sum_{k\lambda} \frac{\varepsilon_k\lambda,i\varepsilon_k\lambda,j}{v} \cos(k \cdot x) \rightarrow \frac{\varepsilon_i\varepsilon_j}{v} \cos(k \cdot x). \tag{124}
\]

With this the \( \alpha \)-gauge transverse material polarisation becomes

\[
P_{T\alpha}(x) = \frac{\alpha}{v} \sum_{\mu=1}^N \varepsilon(d_{\mu} \cdot \varepsilon) \cos[k \cdot (x - R_{\mu})]. \tag{125}
\]

Similarly, \( V_{\text{dip}} \) becomes

\[
V_{\text{dip}} = -\frac{1}{2} \sum_{\mu \neq \nu} \frac{N}{d_{\mu,i}d_{\nu,j}\delta_T^{ij}(R_{\mu} - R_{\nu})} = -\frac{1}{2v} \sum_{\mu \neq \nu} (d_{\mu} \cdot \varepsilon)(d_{\nu} \cdot \varepsilon) \cos[k \cdot (R_{\mu} - R_{\nu})]. \tag{126}
\]

Altogether, within the single-mode theory the Hamiltonian in Eq. (114) becomes

\[
H = \sum_{\mu=1}^N \frac{1}{2m} [p_{\mu\alpha} + \varepsilon(1 - \alpha)A_T(R_{\mu})]^2 + V + \frac{\alpha^2}{2v} \sum_{\mu=1}^N (d_{\mu} \cdot \varepsilon)^2 + (1 - \alpha^2) V_{\text{dip}} + \alpha \sum_{\mu=1}^N d_{\mu} \cdot \Pi_{T\alpha}(R_{\mu}) + \omega \left( a_{\alpha}^\dagger a_{\alpha} + \frac{1}{2} \right). \tag{127}
\]

where \( A_T(R_{\mu}), \Pi_{T\alpha}(R_{\mu}) \) and \( V_{\text{dip}} \) are given by Eqs. (121), (122) and (126) respectively.

Within the single-mode restriction the Heisenberg equation with the Hamiltonian in Eq. (127) yields

\[
E_T(x) = -\dot{A}_T(x) = -i\omega g\varepsilon \left( a_{\alpha}^\dagger e^{-ik\cdot x} - a_{\alpha} e^{ik\cdot x} \right) - \frac{\alpha}{v} \sum_{\mu=1}^N \varepsilon(d_{\mu} \cdot \varepsilon) \cos[k \cdot (x - R_{\mu})] = -\Pi_{T\alpha}(x) - P_{T\alpha}(x). \tag{128}
\]

as required according to Eq. (5). Because the single-mode restriction has been imposed on both the mode operators and the material transverse polarisation the fundamental kinematic relations of the Hamiltonian theory, namely Eqs. (4) and (5), are preserved. We therefore obtain a self-consistent theory describing \( N \) dipoles and a single-mode of radiation. We note that as in the multi-mode theory of Eq. (114) electrostatic inter-dipole interactions are explicit in Eq. (127) in all gauges other than the multipolar gauge \( \alpha = 1 \).

Finally we consider the limit of closely spaced dipoles around the origin \( 0 \) such that \( R_{\mu} \approx 0 \). In this case, according to Eqs. (125) and (126) we obtain

\[
V_{\text{dip}} = -\frac{1}{2v} \sum_{\mu \neq \nu} (d_{\mu} \cdot \varepsilon)(d_{\nu} \cdot \varepsilon), \tag{129}
\]

\[
P_{T\alpha} := P_{T\alpha}(0) = \frac{\alpha}{v} \sum_{\mu=1}^N \varepsilon(d_{\mu} \cdot \varepsilon). \tag{130}
\]

The canonical fields \( A_T(R_{\mu}) \) and \( \Pi_{\alpha}(R_{\mu}) \) within the Hamiltonian are replaced by \( A := A_T(0) \) and \( \Pi_{\alpha} := \Pi_{T\alpha}(0) \) respectively, where for notational convenience we have dropped the transversality subscript \( _T \). We also adopt this convention in the main text. The Hamiltonian in Eq. (127) is now given by Eq. (8) in the main text.
Supplementary Note 2: Diagonalisation of generic bilinear coupled-oscillator Hamiltonian

Here we diagonalise a coupled oscillator Hamiltonian with the generic structure that we will repeatedly encounter. We define arbitrary oscillator operators \( \{ y, y^\dagger, z, z^\dagger \} \) with \([y, y^\dagger] = 1 = [z, z^\dagger] \) and where all other commutators between elements of \( \{ y, y^\dagger, z, z^\dagger \} \) vanish. The generic Hamiltonian we wish to diagonalise is

\[
h := wy^\dagger y + w'z^\dagger z + ig(y^\dagger + y)(z^\dagger - z') - ig'(y^\dagger - y)(z^\dagger + z) + C
\]  

(131)

where \( C \) is a constant. As an example, the Hamiltonian in Eq. (143), which describes the thermodynamic limit of the \( \alpha \)-gauge Dicke model in the normal phase, has the form of \( H \) above.

To diagonalise \( h \) we introduce Hermitian quadratures \( q_\mu = (\mu^\dagger + \mu)/\sqrt{2} \) and \( p_\mu = i(\mu^\dagger - \mu)/\sqrt{2} \) where \( \mu = y, z \). Subsequently we define the tuple of quadratures \( \mathbf{r} = (q_y, q_z, p_y, p_z) \), which is such that \([r_j, r_k] = i\Omega_{jk}\) where

\[
\Omega = \begin{pmatrix} 0 & I \\ -I & 0 \end{pmatrix}
\]  

(132)

is a matrix representation of the standard symplectic form on \( \mathbb{R}^{2\times 2} \). The Hamiltonian in Eq. (131) can now be written

\[
h = \mathbf{r}^T M \mathbf{r} + C - \frac{1}{2}(w + w')
\]  

(133)

where \( \cdot^T \) denotes transposition and

\[
M = \frac{1}{2} \begin{pmatrix} w & 0 & 0 & 2g \\ 0 & w' & -2g' & 0 \\ 0 & -2g' & w & 0 \\ 2g & 0 & 0 & w' \end{pmatrix}
\]  

(134)

is assumed to be positive-definite. By Williamson’s theorem [52] there exists a symplectic matrix \( \Lambda \) such that

\[
\Lambda^T M \Lambda = \tilde{D} = \begin{pmatrix} D & 0 \\ 0 & D \end{pmatrix}
\]  

(135)

where \( D \) is diagonal. Denoting the elements of \( D \) by \( \nu_j, j = 1, 2 \), the quantity \( \pm i\nu_j \) is an eigenvalue of \( \Omega M \). We therefore make use of the canonically transformed quadratures \( \mathbf{r}' = (q'_y, q'_z, p'_y, p'_z) = \Lambda^{-1} \mathbf{r} \), which because \( \Lambda \) is symplectic also satisfy \([r'_j, r'_k] = i\Omega_{jk}\). The Hamiltonian \( h \) can now be written in terms of upper and lower polaritons as

\[
h = \mathbf{r'}^T \tilde{D} \mathbf{r'} + C - \frac{1}{2}(w + w') = E_+ y^\dagger y' + E_- z^\dagger z' + \frac{1}{2}(E_+ + E_-) - w - w') + C
\]  

(136)

where \( y', z' \) are bosonic operators defined in terms of the transformed quadratures \( \mathbf{r}' \). They satisfy \([y', y'^\dagger] = 1 = [z', z'^\dagger] \) while all other commutators between elements of \( \{ y', y'^\dagger, z', z'^\dagger \} \) vanish. The energies \( E_{\pm} \) are given by the elements of \( D \) as \( E_{\pm}^2 = (2\nu_\pm)^2 \) and \( \nu_\pm \) are found from the matrix \( \Omega M \), which is found from Eq. (134). Explicitly, the polariton energies \( E_{\pm} \) are given by

\[
2E_{\pm}^2 = 8gg' + w^2 + w'^2 \pm \sqrt{(w^2 - w'^2)^2 + 16(wg' + w'g')wg + w'g').
\]  

(137)

Supplementary Note 3: Derivation of the normal phase Hamiltonian

The \( \alpha \)-gauge Dicke model Hamiltonian is

\[
H^{\alpha,2} = \omega_m J_z^\alpha + \frac{N}{2}(c_0 + c_1) + \frac{\hbar^2}{N} p_\omega (c^\dagger_\alpha c_\alpha + \frac{1}{2}) - \frac{g}{N} (J^+_\alpha + J^-_\alpha)^2
\]  

\[
- i(1 - \alpha)\omega_m a^\dagger \sqrt{\frac{\rho}{2\omega_m N}} (J^+_\alpha - J^-_\alpha)(c^\dagger_\alpha c_\alpha) + i\alpha d^\dagger \sqrt{\frac{\rho\omega_m}{2N}} (J^+_\alpha + J^-_\alpha)(c^\dagger_\alpha - c_\alpha).
\]  

(138)

Substituting

\[
J^-_\alpha = b^\dagger_\alpha \sqrt{N - b^\dagger_\alpha b_\alpha}, \quad J^+_\alpha = (J^+_\alpha)^\dagger
\]  

(139)
into Eq. (138) gives

\[ H_{\text{th}}^{\alpha,2} = \omega_m \left( b_\alpha^\dagger b_\alpha - \frac{N}{2} \right) + \frac{N}{2} (\epsilon_0 + \epsilon_1) + \frac{1}{2} \rho d^2 + \omega_\alpha \left( c_\alpha^\dagger c_\alpha + \frac{1}{2} \right) - C_\alpha \left( b_\alpha^\dagger \sqrt{1 - \rho \frac{b_\alpha^\dagger b_\alpha}{N}} + \sqrt{1 - \rho \frac{b_\alpha^\dagger b_\alpha}{N}} b_\alpha \right)^2 \]

\[ - i(1 - \alpha) \omega_m a_d \sqrt{\frac{\rho}{2 \omega_\alpha}} \left( b_\alpha^\dagger \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} - \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} b_\alpha \right) (c_\alpha^\dagger + c_\alpha) \]

\[ + i \alpha d \sqrt{\frac{\rho \omega_\alpha}{2}} \left( b_\alpha^\dagger \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} + \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} b_\alpha \right) (c_\alpha^\dagger - c_\alpha). \]  

(140)

All terms that depend on the square-root functions of the mode operators \( b_\alpha, b_\alpha^\dagger \) have coefficients that remain finite in the thermodynamic limit. Therefore, expanding the square-roots as

\[ \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} = 1 - \frac{b_\alpha^\dagger b_\alpha}{2N} + ... \]

(141)

and ignoring terms which vanish in the thermodynamic limit \((N \to \infty)\) constitutes making the replacement

\[ \sqrt{1 - \frac{b_\alpha^\dagger b_\alpha}{N}} \to 1 \]  

(142)

in Eq. (140), which yields

\[ H_{\text{th}}^{\alpha,2} = \omega_m \left( b_\alpha^\dagger b_\alpha - \frac{N}{2} \right) + \frac{N}{2} (\epsilon_0 + \epsilon_1) + \frac{1}{2} \rho d^2 + \omega_\alpha \left( c_\alpha^\dagger c_\alpha + \frac{1}{2} \right) - C_\alpha \left( b_\alpha^\dagger + b_\alpha \right)^2 \]

\[ - i(1 - \alpha) \omega_m a_d \sqrt{\frac{\rho}{2 \omega_\alpha}} (b_\alpha^\dagger - b_\alpha) (c_\alpha^\dagger + c_\alpha) + i \alpha d \sqrt{\frac{\rho \omega_\alpha}{2}} (b_\alpha^\dagger + b_\alpha) (c_\alpha^\dagger - c_\alpha). \]  

(143)

We now combine the terms that depend only on \( b_\alpha, b_\alpha^\dagger \) via a Bogoliubov transformation such that

\[ \omega_m b_\alpha^\dagger b_\alpha - C_\alpha [b_\alpha^\dagger + b_\alpha]^2 = \tilde{\omega}_m \alpha f_\alpha f_\alpha + \frac{1}{2} (\tilde{\omega}_m^\alpha - \omega_m) \]  

(144)

where \([f_\alpha, f_\alpha^\dagger] = 1\) and

\[ \tilde{\omega}_m^\alpha = \omega_m^2 - 4C_\alpha. \]  

(145)

The quadrature operators defined in terms of the \( b_\alpha \) are given in terms of the \( f_\alpha \) by

\[ b_\alpha^\dagger - b_\alpha = \frac{\tilde{\omega}_m^\alpha}{\omega_m} (f_\alpha^\dagger - f_\alpha), \]

(146)

\[ b_\alpha^\dagger + b_\alpha = \frac{\omega_m}{\tilde{\omega}_m^\alpha} (f_\alpha^\dagger + f_\alpha). \]

(147)

Substituting these expressions into Eq. (143) gives

\[ H_{\text{th}}^{\alpha,2} = \tilde{\omega}_m^\alpha f_\alpha^\dagger f_\alpha + \omega_\alpha c_\alpha^\dagger c_\alpha + C_\alpha - i(1 - \alpha) d_a \sqrt{\frac{\omega_m \tilde{\omega}_m^\alpha \rho}{2 \omega_\alpha}} (f_\alpha^\dagger - f_\alpha) (c_\alpha^\dagger + c_\alpha) + i \alpha d \sqrt{\frac{\rho \omega_\alpha}{2 \omega_m}} (f_\alpha^\dagger + f_\alpha) (c_\alpha^\dagger - c_\alpha) \]

(148)

where we have combined all constant terms into

\[ C_\alpha = N \epsilon_0 + \frac{1}{2} \left( \tilde{\omega}_m^\alpha - \omega_m + \omega_\alpha + \rho d^2 \right). \]  

(149)

Note that if we choose the material potential such that \( \epsilon_0 = 0 \), i.e., if the material energy zero-point is zero, then \( C_\alpha \) is \( N \)-independent and remains finite in the limit \( N \to \infty \). The Hamiltonian in Eq. (148) can be diagonalised using the method in Supplementary Note 2, which leads to the final result given in Eq. (32) of the main text.
Supplementary Note 4: Derivation of the abnormal phase Hamiltonian

The relevant starting Hamiltonian $H^{α,2}$ is again that of Eq. (138), together with the Holstein-Primakoff representation

\[ J_α^+ = b_α^† b_α - \frac{N}{2}, \quad J_α^- = \sqrt{N - b_α^† b_α}, \quad J_α^z = (J_α^+)^†. \]  

(150)

We then introduce the displaced operator $f_α = b_α - \sqrt{β_α}$ where $β_α$ is assumed to be of order $N$, such that the material part $H_m^{α,2}$ of $H^{α,2}$ can be written

\[ H_m^{α,2} = \omega_m b_α^† b_α - \frac{C_α}{N} \left( b_α^† \sqrt{N - b_α^† b_α} + \sqrt{N - b_α^† b_α} b_α \right)^2 \]

\[ = \omega_m \left( f_α^† f_α - \sqrt{β_α} [f_α^† f_α] + β_α \right) - C_α \frac{N - β_α}{N} \left( [f_α^† - \sqrt{β_α}] \sqrt{ξ_α} + \sqrt{ξ_α} [f_α - \sqrt{β_α}] \right)^2 \]  

(151)

where for convenience we have defined

\[ ξ_α := 1 - \frac{f_α^† f_α - \sqrt{β_α} [f_α^† f_α]}{N - β_α}. \]  

(152)

Expanding $\sqrt{ξ_α}$ in Eq. (151) and neglecting terms which vanish in the thermodynamic limit one obtains after lengthy manipulations

\[ H_{th,m}^{α,2} = (\omega_μ + 4C_α \frac{β_α}{N}) f_α^† f_α - \frac{C_α}{N} (N - 5β_α) (f_α^† + f_α)^2 + β_α ω_m - 2C_α \left( \frac{N - β_α}{N} \right) \left( 2β_α + \frac{β_α}{N - β_α} \right) \]

\[ - \sqrt{β_α} \left( ω_m - 4C_α \frac{N - 2β_α}{N} \right) (f_α^† + f_α). \]  

(153)

Next we replace the radiation mode operators with displaced operators such that $c_α = i(k_α + \sqrt{γ_α})$. The total Hamiltonian therefore reads

\[ H_{th}^{α,2} = H_{th,m}^{α,2} + ω_α \left( k_α^† k_α + \sqrt{γ_α} [k_α^† + k_α] + γ_α + \frac{1}{2} \right) - g_α \sqrt{\frac{N - β_α}{N}} (f_α^† \sqrt{ξ_α} - \sqrt{ξ_α} f_α) (k_α^† - k_α) \]

\[ + g_α \sqrt{\frac{N - β_α}{N}} (f_α^† \sqrt{ξ_α} + \sqrt{ξ_α} f_α - 2\sqrt{β_α} ξ_α) (k_α^† + k_α + 2\sqrt{γ_α}) + \frac{1}{2} ρ d^2 + N ε_0 \]  

(154)

We now collect all terms that are linear in the $f_α$ or in the $k_α$ and choose $β_α$ and $γ_α$ such that these terms vanish. The trivial case in which $β_α = 0 = γ_α$ yields the normal phase Hamiltonian. We will see that the non-trivial solutions

\[ β_α = β := \frac{N}{2} (1 - τ), \]  

(155)

\[ γ_α = \frac{N g_α^2}{ω_α^2} (1 - τ^2) \]  

(156)

where

\[ τ := \frac{ω_μ ω_m}{4 (g_α^2 + ω_α C_α)} = \frac{α^2 ω_α ω_m}{4 g_α^2} = \frac{ω_m}{2 ρ d^2}. \]  

(157)

yield a Hamiltonian describing the abnormal phase.

Expanding $\sqrt{ξ_α}$ and neglecting terms which vanish in the thermodynamic limit one now obtains after lengthy
We can remove the term quadratic in \( f_a^\dagger + f_a \) by defining new material mode operators \( f'_\alpha, f'^\dagger_\alpha \) such that

\[
\begin{align*}
\left( \omega_m + 4\alpha \frac{\beta}{N} + 2g\alpha \sqrt{\frac{\gamma\alpha\beta}{N(N - \beta)}} \right) f'_\alpha f_\alpha + \frac{1}{2} \right) \\
+ \left( \sqrt{\frac{\gamma\alpha\beta}{N(N - \beta)}} \left[ 1 + \frac{\beta}{2(N - \beta)} - \frac{\alpha}{N} (N - 5\beta) \right] \frac{f'^\dagger_\alpha f_\alpha}{N} \right) f'^\dagger_\alpha f_\alpha + \frac{1}{2} \\
+ g\alpha \sqrt{\frac{N - \beta}{N}} \left( 1 - \frac{\beta}{N - \beta} \right) (f'_\alpha f_\alpha + f_\alpha f'^\dagger_\alpha) \\
+ N\epsilon_0 + \frac{1}{2} \rho d^2 + \frac{\omega_m}{2} + \gamma\alpha\omega_\alpha + 2\gamma\alpha\beta (1 + 2[N - \beta]) - g\alpha \sqrt{\frac{\gamma\alpha\beta}{N(N - \beta)}} (1 + 4(N - \beta))
\end{align*}
\]

We can remove the term quadratic in \( f'_\alpha + f_\alpha \) by defining new material mode operators \( f'^\dagger_\alpha, f'_\alpha \) such that

\[
\begin{align*}
\left( \omega_m + 4\alpha \frac{\beta}{N} + 2g\alpha \sqrt{\frac{\gamma\alpha\beta}{N(N - \beta)}} \right) f'^\dagger_\alpha f_\alpha + \frac{1}{2} \\
+ \left( \sqrt{\frac{\gamma\alpha\beta}{N(N - \beta)}} \left[ 1 + \frac{\beta}{2(N - \beta)} - \frac{\alpha}{N} (N - 5\beta) \right] \frac{f'^\dagger_\alpha f_\alpha}{N} \right) f'^\dagger_\alpha f_\alpha + \frac{1}{2} \\
= \frac{\omega_m}{2\tau} (1 + \tau) \left( f'^\dagger_\alpha f_\alpha + \frac{1}{2} \right) + \left( \frac{\alpha}{2} [3 - 5\tau] + \frac{\omega_m\alpha^2(1 - \tau)(3 + \tau)}{8\tau(1 + \tau)} \right) \frac{f'^\dagger_\alpha f_\alpha}{N}
\end{align*}
\]

where

\[
\omega'^2 = \frac{\omega_m^2}{\tau^2} \left[ 1 - (1 - \alpha^2)^2 \right].
\]

Letting \( \omega'^m = \omega_m(1 + \tau)/(2\tau) \) and using the relations

\[
\begin{align*}
f'^\dagger_\alpha - f_\alpha &= \frac{\omega'^m}{\omega_m} (f'^\dagger_\alpha - f'^\dagger_\alpha) \\
f'^\dagger_\alpha + f_\alpha &= \frac{\omega'_m}{\omega_m} (f'^\dagger_\alpha + f'^\dagger_\alpha)
\end{align*}
\]

the Hamiltonian can be written

\[
H^\alpha_{\text{th}} = \omega'_m f'^\dagger_\alpha f'_\alpha + \omega_\alpha c'^\dagger_\alpha c'_\alpha - ig'_\alpha (f'^\dagger_\alpha - f'^\dagger_\alpha) (c'^\dagger_\alpha + c'_\alpha) + ig_\alpha (f'^\dagger_\alpha + f'^\dagger_\alpha) (c'^\dagger_\alpha - c'_\alpha) + C_\alpha
\]

where \( c'_\alpha = ik_\alpha \) while

\[
\begin{align*}
g'_\alpha &= \sqrt{\frac{\tau\omega'^m}{\omega_m}} g_\alpha, \\
g_\alpha &= \sqrt{\frac{\tau\omega'_m}{\omega_m}} g_\alpha
\end{align*}
\]

and

\[
C'_\alpha = N \left( \epsilon_0 - \frac{g'^2_\alpha}{\omega'_m \omega_\alpha} [1 - \tau^2] + \frac{\omega'_m}{2} [1 - \tau^2] - \frac{g'^2_\alpha}{\omega'_m \omega_\alpha} (1 - \tau) + \frac{\omega_m}{2} \right) + \frac{\omega_m}{2} - \frac{\omega'_m}{2} + \frac{1}{2} \rho d^2.
\]

The Hamiltonian in Eq. (162) has the form in Eq. (131) and can therefore be diagonalised by the method in Supplementary Note 2, leading to the final result denoted \( H^\alpha_{2,\text{th}} \), and given in Eq. (46) of the main text.
Supplementary Note 5: Calculation of radiative canonical operator averages

We begin with the cavity canonical operators $A = \varepsilon \cdot A$ and $\Pi_\alpha = \varepsilon \cdot \Pi_\alpha$. The ground state average of $A$ is trivially zero. Similarly the ground state average of $\Pi_\alpha$ in the normal phase is zero. The ground state average of $\Pi_\alpha$ in the abnormal phase can be calculated using the Dicke-model of any gauge $\alpha'$. We begin with the expression

$$\Pi_{\alpha'}^{\alpha,2} = \Pi_{\alpha'} - \frac{d}{v}(\alpha - \alpha')(J_{\alpha'}^+ + J_{\alpha'}^-),$$

which is the $\alpha'$-gauge's two-level approximation of $\Pi_\alpha$. Using the Holstein-Primakoff representation and then defining the displaced operators $f_\alpha$, $c_\alpha$ by

$$b_\alpha = f_\alpha - \sqrt{\beta},$$
$$c_\alpha = c_\alpha' + i\sqrt{\gamma},$$

one obtains

$$\Pi_{\alpha'}^{\alpha,2} = \sqrt{\frac{2\omega_\alpha}{N}} (i\left[c_{\alpha'}' - c_{\alpha'}'\right] + 2\sqrt{\gamma}) - \frac{d}{v}(\alpha - \alpha')\sqrt{\frac{N}{\beta}} \left(f_{\alpha'}'\sqrt{\xi_{\alpha'}}' + \sqrt{\gamma}\xi_{\alpha'}f_{\alpha'}' - 2\sqrt{\beta}\xi_{\alpha'}'\right)$$

where $\beta$, $\gamma_\alpha$, and $\xi_\alpha$ are defined in Eqs. (155), (156), and (152) respectively. Expanding $\sqrt{\xi_{\alpha'}}$ and retaining terms which do not vanish in the thermodynamic limit yields

$$\Pi_{\alpha',\text{th}}^{\alpha,2} = \sqrt{\frac{2\omega_\alpha\gamma_{\alpha'}}{N}} + 2(\alpha - \alpha')d\sqrt{(N - \beta)/v} = \alpha d\sqrt{1 - \tau^2},$$

as given in Eq. (62) in the main text.

Supplementary Note 6: Alternative implementations of the two-level truncation

Here we consider alternatives to the two-level truncation defined in Sec. III A. The adopted conventions for the two-level truncation were shown to yield accurate predictions in Sec. V. If in the same example the two-level Hamiltonian is defined by the right-hand-side of Eq. (26), that is, by $\tilde{H}^{2,\alpha} = HP_\alpha HP_\alpha$ then accurate predictions are also obtained. The only non-linear material interaction term in the Hamiltonian is the polarisation self-interaction term, from which it follows that for the example considered

$$P_\alpha = \sum_{\mu=1}^{N} \frac{\langle \varepsilon \cdot d \rangle^2}{2v} \approx \sum_{\mu=1}^{N} \frac{\langle \varepsilon \cdot d \rangle^2}{2v}. \quad (171)$$

Of greater interest is the possibility of including the polarisation self-energy within the definition of the material bare energy as

$$\hat{H}_m^\alpha = \sum_{\mu=1}^{N} \frac{P_{\mu}^2}{2m} + V + \frac{a^2}{2} \sum_{\mu=1}^{N} (d_{\mu} \cdot \varepsilon)^2 = \sum_{\mu=1}^{N} \sum_{n=0}^{\infty} \epsilon_n \left| \langle \epsilon_{\mu,n}^\alpha \rangle \right| \approx \omega_m \sum_{\mu=1}^{N} \sigma_{\mu}^2 + N \left( \epsilon_0^\alpha + \epsilon_1^\alpha \right) \quad (172)$$

where now the eigenstates $|\epsilon_{\mu,n}^\alpha\rangle$, eigenenergies $\epsilon_n^\alpha$, and transition dipole moment $d_\alpha = \langle \epsilon_{\mu,0}^\alpha | d_{\mu} | \epsilon_{\mu,1}^\alpha \rangle$ are all $\alpha$-dependent. This convention for the two-level truncation was considered in Ref. [41] for example. In the case of a single double-well dipole we have

$$\hat{H}_m^\alpha = \frac{p_a^2}{2m} + \frac{1}{2} \left( \rho a^2 e^2 - \theta \right) r^2 + \frac{\phi}{4} r^4$$

$$= \frac{\varepsilon}{2} \left( -\partial^2_{\xi} + \left( \frac{\Omega \eta}{\varepsilon} \right)^2 - \beta \right) \xi^2 + \frac{\xi^4}{2} \quad (173)$$

where $\rho = 1/v$, because $N = 1$. In this case $\omega_m^\alpha$ and $d_\alpha$ can be obtained numerically and viewed as functions of both $\alpha$ and $\eta$. However, in taking the thermodynamic limit $N \to \infty$, implying that $N \neq 1$. Instead, the ratio $\rho = N/v$ is
FIG. 4: In all plots we have chosen $\beta = 2.4$, and then chosen $E$ such that $\omega_m^0 = \omega_m = \omega$, that is, we set parameters such that the bare cavity frequency is resonant with the Coulomb-gauge material transition frequency, which is coupling-independent, because there is no polarisation self-energy term in the Coulomb gauge. (a) The lower polariton energy is plotted for three values of $\alpha$ as a function of $\eta$, with the inset showing a zoom around the phase transition point, which is now different depending on the value of $\alpha$. The qualitative behaviour of $E_{\alpha-}$ as $\eta$ becomes large depends on the value of $\alpha$. (b) The upper polariton energy is plotted for three values of $\alpha$ as a function of $\eta$. As with the lower polariton energy the behaviour for large $\eta$ depends on the value of $\alpha$.

Supplementary Note 7: Further numerical results

Here we consider a less anharmonic single-dipole double-well potential, which has $(\epsilon_2 - \epsilon_0)/\omega_m \approx 3.2$. This results from choosing $\beta = 1.5$ rather than $\beta = 3.3$ as was chosen in the main text. In this case single-dipole two-level models are able to remain accurate in predicting the low energy properties of the system, but the multipolar gauge no longer provides the optimal two-level model. The optimal gauge for the two-level truncation is shifted towards the Coulomb gauge, such that the Jaynes-Cummings gauge two-level model is close to optimal [43]. This is shown in Fig. 6, which compares $G$ (Fig. 6a), $E - G$ (Fig. 6b), and $d^2G/d\eta^2$ (Fig. 6c) each obtained from the Coulomb-gauge, Jaynes-Cummings gauge, multipolar-gauge two-level models, and the exact (non-truncated) theory. For $N > 1$, two-level models become less accurate in predicting even low energy properties when the coupling is sufficiently strong, as
FIG. 5: The quantity $d_\alpha' \Pi_{\alpha,\text{th}}^{\alpha',2}$ is plotted as a function of $\eta$ for $\alpha \in \{0.5, 1\}$ and $\alpha' \in \{0, 0.5, 1\}$. As in Fig. 4, we have chosen $\beta = 2.4$, and then chosen $\beta$ such that $\omega^0_m = \omega$. As in Fig. 4, the phase transition location is different depending on the chosen gauge $\alpha'$ in which the two-level truncation is performed. The definition of the canonical momentum changes linearly with $\alpha$ from $\alpha = 0$ such that $\Pi_0 = -E_T$ to $\alpha = 1$ such that $\Pi_1 = -D_T$. Correspondingly, for fixed $\alpha'$ we have $d_\alpha' \Pi_{\alpha,\text{th}}^{\alpha',2}/d_\alpha' \Pi_{\alpha',\text{th}}^{\alpha',2} = \alpha/\alpha'$. Thus, the ratio of the magnitude of each solid line to the magnitude of the dashed line with the same colour is constant, with value $0.5/1 = 0.5$. Note in addition, that $d_\alpha' \Pi_{\alpha,\text{th}}^{\alpha',2} = d_\alpha' P_{\alpha,\text{th}}^{\alpha',2}$, meaning that the dashed curves corresponding to $\alpha = 1$ illustrate the different $\alpha'$-gauge two-level approximations of the gauge-invariant manifestation of the abnormal phase via the transverse polarisation $P_T$.

shown for the case $N = 2$ in Fig. 7 and for the case $N = 3$ in Fig. 8. The accuracy of multipolar-gauge two-level truncation appears to improve relative to the other gauges, but the ground energy is not well represented by any two-level model for sufficiently strong coupling.
FIG. 6: In all plots we have chosen $\beta = 1.5$ and then chosen $\mathcal{E}$ such that $\omega_m = \omega$. The single-dipole Coulomb gauge, Jaynes-Cummings gauge and multipolar gauge two-level model predictions are compared with the corresponding exact predictions as a function of $\eta$ for: (a) the ground energy $G$, (b) the first transition energy $E - G$, (c) the second derivative $d^2G/d\eta^2$. In all cases the Jaynes-Cummings gauge two-level model is most accurate.
FIG. 7: In all plots we have chosen $\beta = 1.5$ and then chosen $\mathcal{E}$ such that $\omega_m = \omega$. For the two-dipole case ($N = 2$), the Coulomb gauge, Jaynes-Cummings gauge and multipolar gauge two-level model predictions are compared with the corresponding exact predictions as a function of $\eta$ for: (a) the ground energy $G$, (b) the first transition energy $E - G$.

FIG. 8: In all plots we have chosen $\beta = 1.5$ and then chosen $\mathcal{E}$ such that $\omega_m = \omega$. For the three-dipole case ($N = 3$), the Coulomb gauge, Jaynes-Cummings gauge and multipolar gauge two-level model predictions are compared with the corresponding exact predictions as a function of $\eta$ for: (a) the ground energy $G$, (b) the first transition energy $E - G$. 