Master's Thesis in Physics

Photon condensation in magnetic cavity QED

Juan Román Roche

Director:
David Zueco Láinez

Universidad Zaragoza 1542

Facultad de Ciencias Universidad Zaragoza

Department of Condensed Matter Physics
Faculty of Science, University of Zaragoza

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Introduction

Bose-Einstein condensation constitutes a cornerstone of physics. It is explained in every statistical mechanics course as a clear manifestation of the intrinsic difference between bosons and fermions. Hence, it is not surprising that its 1995 experimental verification [1] deserved a Nobel prize, 70 years after the famous *collaboration* between Bose and Einstein. Ever since, low temperature laboratories around the world have carried out innumerable experiments revolving around this phenomenon. Very recently, even the International Space Station achieved Bose-Einstein condensation [2]. Yet, physicists have failed to achieve Bose-Einstein condensation of photons, ironically the most abundant boson in our universe. The difficulty stems from the fact that photons are massless, and thus not subject to conservation laws as massive bosons are. When cooled to condensation temperatures, photons are absorbed by the surrounding matter, instead of macroscopically populating the ground state, as is characteristic of Bose-Einstein condensation. A solution to condense photons is to consider hybrid light-matter quasiparticles known as polaritons, which work by dressing the photons with an effective mass [3]. Here, we explore a different alternative.

More than 47 years ago Hepp and Lieb showed that *photon condensation* was theoretically possible in Dicke's model [4]. In this model, symmetry breaking was induced by the coupling of an electromagnetic cavity to the electric dipoles of N free atoms in the thermodynamic limit $N \to \infty$ (See Fig. 1). The experimental realization of this model has been pursued for the last 47 years [5]. However, the transition has never been measured. During this time, the community has enjoyed a tortuous succession of proposals on how to achieve photon condensation, each shortly matched with a corresponding no-go theorem [6–13].

In this Master's Thesis we present a no-go theorem that unifies all these no-go theorems (including some recent ones) and we propose a rather straightforward way to avoid them: harnessing magnetic coupling. Therefore, we solve this long-standing theoretical controversy and provide a realistic experimental layout to measure the transition, using magnetic molecules (instead of electric-dipole-coupled ones).

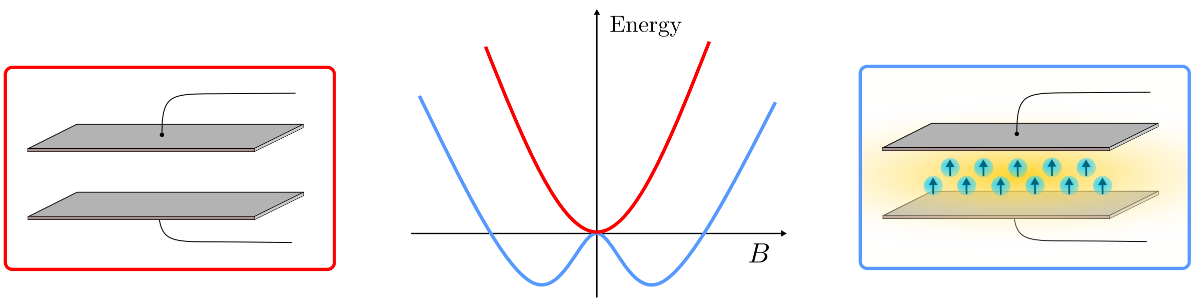

Fig. 1: Schematic representation of the spontaneous symmetry breaking induced by the addition of dipoles to an electromagnetic cavity. This gives rise to a finite value of the magnetic field B in the cavity (or equivalently to the electric field E) which implies that there is a population of photons in the cavity at equilibrium.

Objectives and outline

The work presented in this Master's Thesis has followed a series of objectives. Some were laid down at the beginning of the project, while others have been added as our understanding of the problem deepened. Nevertheless, we list them here indistinctly. First, to understand the historical developments in the study of photon condensation, addressing the relevant literature, which spans more that 47 years of contributions to the topic [4]. Our intention was to get a grasp of the state of affairs of the field, allowing us to disentangle the back and forth of no-go and counter no-go theorems. In that sense, our aim was to generate some intuition on the issue and provide a holistic interpretation, possibly providing some definitive no-go theorem. Simultaneously, we would explore the possibility of achieving photon condensation by exploiting magnetic coupling. This is motivated by the fact that from the beginning of the project, specially since what got us started was Andolina and colleagues' no-go theorem [12], we were aware that the evidence supporting the impossibility of superradiance using the coupling to the electric dipole was strong. Exploring magnetic cavity QED avoided the existing no-go theorems and any possible no-go theorem that we might end up contributing as a consequence of our advances in our first objectives. We sought to provide a full characterization of the model, studying the values of the physical parameters at which the transition occurs and a broad range of generalizations, in order to gauge its experimental feasibility. Consequently, our work would finish with a proposal of an experiment to measure the superradiant phase transition, catering to the possibilities and know-how of the experimentalists in our research group (QMAD), which have a track record in coupling magnetic molecules to CPW resonators.

Consequently, this Master's Thesis is divided in two blocks. The first block discusses the problem of photon condensation as presented in the literature, with electric-dipole coupling. Section 1 starts with a brief presentation of Pauli's equation and how it leads to the Hamiltonian of the model under study, we then proceed to give a thorough overview of the historical contributions to the topic, from Hepp and Lieb's original contribution to the present day. Then, in Sec. 2, we present a unified no-go theorem that settles the debate, proving that *photon condensation does not occur when the coupling between light and matter is through the electric dipole*. The second block explores magnetic cavity QED. In Sec. 3 we introduce Zeeman coupling in our Hamiltonian of the model while considering molecules without electric dipole, we show that this leads to the Dicke model, in which superradiance occurs. After finding that magnetic cavity QED permits photon condensation we test the robustness of the model against some generalizations. Following the success in Sec. 3, in Sec. 4 we discuss a transmission experiment designed to measure the phase transition. Finally, we draw some conclusions from our results and outline possible continuations. Technical details are left for the appendices.

1 Photon condensation: a historical perspective

1.1 From Dirac to cavity QED

The Pauli equation is the non-relativistic limit of the Dirac equation. It describes the interaction of non-relativistic spin 1/2 particles with an electromagnetic field [14, Chap. 2.1].

$$\mathcal{H}_P = \frac{1}{2m}\left(\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}\right)^2 + e\phi - \frac{e\hbar}{2mc}\boldsymbol{\sigma}\boldsymbol{H} + \zeta\boldsymbol{l}\boldsymbol{\sigma} \tag{1}$$

From left to right, we have the minimally coupled kinetic energy, the electromagnetic energy, the Zeeman coupling and spin-orbit coupling. We can write the generalization to a system consisting of many electrons

$$\begin{aligned}\mathcal{H} = &\sum_i \frac{\left(\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}(\boldsymbol{r}_i)\right)^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + e\phi \\ &- \sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}_i\boldsymbol{B}(\boldsymbol{r}_i) + \sum_i \zeta\boldsymbol{l}_i\boldsymbol{\sigma}_i.\end{aligned} \tag{2}$$

If we consider the source of the electromagnetic field to be an optical or superconducting cavity, we quantize the electromagnetic field in the Coulomb gauge, where $\boldsymbol{A}(\boldsymbol{r}_i) = \sum_l^M \boldsymbol{A}_l(\boldsymbol{r}_i)\left(a_l + a_l^\dagger\right)$, $\boldsymbol{B}(\boldsymbol{r}_i) = \boldsymbol{\nabla}\times\boldsymbol{A}(\boldsymbol{r}_i)$, $e\phi = \sum_l^M \hbar\omega_l a_l^\dagger a_l$, and $a, a^\dagger$ are the bosonic annihilation and creation operators. In most instances, we will consider only the fundamental mode of the cavity, so the quantized Hamiltonian in the Coulomb gauge reads

$$\begin{aligned}\mathcal{H} = &\sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c a^\dagger a + \sum_i \frac{e}{mc}\boldsymbol{p}_i\boldsymbol{A}_0(\boldsymbol{r}_i)\left(a + a^\dagger\right) \\ &+ \sum_i \frac{e^2\boldsymbol{A}_0^2(\boldsymbol{r}_i)}{2mc^2}\left(a + a^\dagger\right)^2 - \sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}_i\boldsymbol{B}_0(\boldsymbol{r}_i)\left(a + a^\dagger\right) + \sum_i \zeta\boldsymbol{l}_i\boldsymbol{\sigma}_i.\end{aligned} \tag{3}$$

1.2 The Dicke model: Hepp and Lieb's prediction

In quantum optics and condensed matter physics, one often seeks a simple, solvable Hamiltonian that captures a specific feature of a system. The Dicke model is obtained from Hamiltonian (3) through a series of assumptions: (a) The long wavelength limit implies the vector potential can be considered uniform in the region of the cavity populated with atoms, $\boldsymbol{A}_0(\boldsymbol{r}_i) \approx \boldsymbol{A}_0(0) = \boldsymbol{A}_0$; (b) the $\boldsymbol{A}_0^2$, Zeeman and spin-orbit terms are neglected; (c) the single electron energy levels can be restricted to only two levels whose transition frequency ω_z is the only one sufficiently close to resonance with the cavity (Two Level Approximation). These approximations yield the Hamiltonian for the Dicke model

$$\mathcal{H} = \omega_c a^\dagger a + \frac{\omega_z}{2}\sum_j \sigma_j^z + \frac{\lambda}{\sqrt{N}}\sum_j \sigma_j^x\left(a + a^\dagger\right). \tag{4}$$

Where σ^μ are the Pauli matrices. Note that we have set $\hbar = 1$ for convenience, we might reintroduce it at will when necessary to provide actual values in a proposed experiment. The coupling between the spins and photons arises from the interaction between the electric dipoles and the electric field, i.e. from *electric coupling*. Consequently [15]

$$\lambda = \frac{d}{\hbar c}\sqrt{\frac{\hbar\omega_c}{2\epsilon_0}\rho}. \tag{5}$$

Where ρ is the electron density and d is the projection of the dipole moment along the cavity field. Dicke first proposed his model for superradiance in 1954 to describe the coherent emission of light by an ensemble of atoms [16]. He was concerned with the *dynamical* process that occurs when the ensemble is prepared with all its constituent atoms, approximated by two-level systems, in their excited state. Left to evolve, one of the atoms will eventually decay, emitting a photon. This in turn triggers a chain reaction that prompts the decay of all remaining atoms. For a system of N atoms we naively expect the emission of proportionally many photons, that is N photons. Dicke showed that for sufficiently packed atoms, within less than a photons' wavelength, the emitted photons would be indistinguishable, interfering constructively to produce an energy density that scales as N^2, in clear contrast with the scenario of independent atoms in which it scaled as N. Such phenomenon was termed superradiance.

Later, in 1973, Hepp and Lieb discovered a different type of transition that occurs in *thermal equilibrium* in the Dicke model [4]. This constitutes a proper quantum phase transition originating from a non-analitic change in the ground state properties and should not be confused with the phenomenon originally described by Dicke. To distinguish them, Hepp and Lieb's discovery was later termed photon condensation, although the community has failed to adhere to the nomenclature in the subsequent literature. In this work we will focus solely on the phenomenon described originally by Hepp and Lieb so, continuing the trend, we will take the liberty to refer to it both as "photon condensation" and as "superradiance". Notice that in order to have a well defined thermodynamic limit, the coupling constant in Hamiltonian (4) scales as $N^{-1/2}$. This is to counter the scaling of the bosonic operators a and $a^\dagger$, which scale as $\sqrt{N}$ (See App. A for proof), leaving the coupling term extensive, i.e. scaling as N. As a consequence, in the normal or "subradiant" phase the number of photons n is zero, while in the superradiant phase n is proportional to N, as will be proven shortly. Hepp and Lieb provided a full proof of photon condensation, valid at finite temperature in the thermodynamic limit. This was later refined by Wang and Hioe that same year, who presented the problem in a less mathematical manner, more amenable to physicists [17] which was later made rigorous by, again, Hepp and Lieb [18]. We will now present the exact solution of the Dicke model, in the thermodynamic limit, combining the contributions made by the aforementioned authors.

It is convenient to consider the basis of coherent states $|\alpha\rangle$ for the photonic mode. Coherent states are defined as the eigenstates of the annihilation operator, such that $a|\alpha\rangle = \alpha|\alpha\rangle$. They form an overcomplete set of states, with closure relation

$$\mathbb{I} = \frac{1}{\pi}\int d^2\alpha \;\; |\alpha\rangle\langle\alpha| \,. \tag{6}$$

The integration is over the complex plane, with $d^2\alpha \; = d\,\mathrm{Re}(\alpha) d\,\mathrm{Im}(\alpha)$. Accordingly, we can write the partition function for the dicke model (4) as

$$Z = Tr\left(e^{-\beta\mathcal{H}}\right) = Tr_S\left(\frac{1}{\pi}\int d^2\alpha \;\; \langle\alpha| e^{-\beta\mathcal{H}} |\alpha\rangle\right). \tag{7}$$

Where $Tr_S(\hat{O})$ is the partial trace over the spin degrees of freedom of operator $\hat{O}$. We do not know in general how to compute $\langle\alpha|\exp[-\beta\mathcal{H}]|\alpha\rangle$, since the expansion of $\exp[-\beta\mathcal{H}]$ would contain terms with disordered combinations of a and $a^\dagger$ of the form $a^\dagger a a a^\dagger \cdots a a^\dagger$. We can, however, and this is a central property that we will use in the remainder of our work, prove certain upper and lower bounds for Z (See App. B.1), these were originally proven by Lieb [19] and Simon [20].

$$\bar{Z} \leq Z \leq e^{\beta\omega_c}\bar{Z}; \qquad \text{with} \qquad \bar{Z} = Tr_S\left(\frac{1}{\pi}\int d^2\alpha\ e^{-\beta\mathcal{H}(\alpha)}\right). \tag{8}$$

Where

$$\mathcal{H}(\alpha) = \langle\alpha|\mathcal{H}|\alpha\rangle = \omega_c|\alpha|^2 + \frac{\omega_z}{2}\sum_j \sigma_j^z + \frac{\lambda}{\sqrt{N}}\sum_j \sigma_j^x(\alpha+\alpha^*). \tag{9}$$

Using Eq. (8) we can write bounds for the free energy per spin

$$-\frac{1}{\beta N}\ln\bar{Z} \geq -\frac{1}{\beta N}\ln Z \geq -\frac{1}{\beta N}\ln\bar{Z} - \frac{\omega_c}{N}, \tag{10}$$

and we find that in the thermodynamic limit $N\to\infty$, $\bar{Z}$ and Z become equivalent. Let us showcase a physical consequence of these bounds. Consider again the expansion of $\exp[-\beta\mathcal{H}]$, we know how a normal-ordered set of bosonic operators acts on a coherent state: $\langle\alpha|(a^\dagger)^m a^n|\alpha\rangle = (\alpha^*)^m\alpha^n$. A general term of the expansion can be expressed as a sum of normal-ordered terms, take for example

$$a^\dagger a a^\dagger a a^\dagger a = a^\dagger a^\dagger a^\dagger a a a + 3\left[a, a^\dagger\right] a^\dagger a^\dagger a a + \left[a, a^\dagger\right]^2 a^\dagger a. \tag{11}$$

The commutator $\left[a, a^\dagger\right]$ equals 1 in the quantum limit, this is derived form the canonical commutation $[x, p] = i\hbar$. However, in the classical limit $[x, p] = 0$, and thus, $\left[a, a^\dagger\right] = 0$. As a consequence, in the classical limit we can drop all terms in the expansion containing a disorderd set of bosonic operators and obtain $\langle\alpha|\exp[-\beta\mathcal{H}]|\alpha\rangle = \exp[-\beta\langle\alpha|\mathcal{H}|\alpha\rangle] = \exp[-\beta\mathcal{H}(\alpha)]$. To sum up, from the bounds for the partition function in Eq. (8) follows that in the thermodynamic limit we can consider the cavity to behave classically. That is, we can consider the bosonic operators a, $a^\dagger$ as c-numbers: α, α^*.

We can now proceed with the calculation of $\bar{Z}$. The spins are independent at this point, so we can write

$$\bar{Z} = \frac{1}{\pi}\int d^2\alpha\ Tr_S\left(e^{-\beta\mathcal{H}(\alpha)}\right) = \frac{1}{\pi}\int d^2\alpha\ e^{-\beta\omega_c\alpha^2}\left(Tr_S e^{-\beta h(\alpha)}\right)^N, \tag{12}$$

with

$$h(\alpha) = \frac{\omega_z}{2}\sigma^z + \frac{\lambda}{\sqrt{N}}\sigma^x(\alpha+\alpha^*) \equiv \begin{pmatrix} \frac{\omega_z}{2} & \frac{\lambda}{\sqrt{N}}(\alpha+\alpha^*) \\ \frac{\lambda}{\sqrt{N}}(\alpha+\alpha^*) & -\frac{\omega_z}{2} \end{pmatrix}. \tag{13}$$

$h(\alpha)$ has eigenvalues $\pm E$, with

$$E = \sqrt{\frac{\omega_z^2}{4} + \frac{4\lambda^2\,\mathrm{Re}(\alpha)^2}{N}}. \tag{14}$$

Hence

$$\begin{aligned}\bar{Z} &= \frac{1}{\pi}\int d^2\alpha\ e^{-\beta\omega_c\alpha^2}\left(2\cosh\beta E\right)^N \\ &= \sqrt{\frac{1}{\pi\beta\omega_c}}\int_{-\infty}^{\infty} d\,\mathrm{Re}(\alpha)\ e^{-\beta\omega_c\,\mathrm{Re}(\alpha)^2}\left\{2\cosh\left(\beta\sqrt{\frac{\omega_z^2}{4} + \frac{4\lambda^2\,\mathrm{Re}(\alpha)^2}{N}}\right)\right\}^N \\ &= \sqrt{\frac{N}{\pi\beta\omega_c}}\int_{-\infty}^{\infty} dz\,\exp\left[-N\left\{\beta\omega_c z^2 - \ln 2\cosh\left(\beta\sqrt{\frac{\omega_z^2}{4} + 4\lambda^2 z^2}\right)\right\}\right]. \end{aligned} \tag{15}$$

We have used the change of variable $z^2 = \mathrm{Re}(\alpha)^2/N$ in the last step. By the saddle point method, in the thermodynamic limit, the only significant contribution to the integral occurs when the exponent is minimum. Minimizing

$$f(z) = \beta\omega_c z^2 - \ln 2\cosh\left(\beta\sqrt{\frac{\omega_z^2}{4} + 4\lambda^2 z^2}\right) \tag{16}$$

with respect to z shows to possible solutions: either $z^* = 0$, corresponding to the subradiant phase; or $\omega_c E - 2\tanh(\beta E)\lambda^2 = 0$, which leads to a finite value of z^* and thus of $\mathrm{Re}(\alpha)^*$. Notice from the calculation of $\bar{Z}$ that $\langle \mathrm{Im}(\alpha)\rangle = 0$, so the only contribution to the average value of α comes from its real part. The saddle point method implies that the true free energy per spin $f = -\ln \bar{Z}/\beta N$ is equal to minimum value of the z-dependent free energy per spin $f(z)$: $f = \min[f(z)]$. As a direct consequence, $\langle z\rangle = z^*$ and so $\langle\alpha\rangle = \alpha^* = \mathrm{Re}(\alpha)^*$. This is all to rigorously justify that we can, at this point, disregard completely the imaginary part of α and refer to both $\langle\alpha\rangle = \mathrm{Re}(\alpha)^*$ as α in the following.

The critical value of λ at which the transition occurs can be obtained form several conditions, one of which is the loss of stability of the $\alpha = 0$ solution, which occurs at

$$\lambda_c = \frac{1}{2}\sqrt{\omega_c\omega_z \coth\left(\beta\frac{\omega_z}{2}\right)}. \tag{17}$$

This is the main result of Hepp and Lieb's 1973 paper [4], and the beginning of the story. The zero temperature limit is $\lambda_c(0) = \frac{1}{2}\sqrt{\omega_c\omega_z}$. Obtaining the value of α in the superradiant phase requires solving the transcendental equation and cannot be done analytically. We can however, expand E in powers of α close to the transition, which provides some insight into the scaling properties of α. The calculation is rather cumbersome, so we skip to the result, a full derivation can be found in App. C.

$$\alpha = \sqrt{N\mathcal{A}(\lambda)\,(\lambda^2 - \lambda_c^2)} \tag{18}$$

where $\mathcal{A}(\lambda)$ is an analytical function of λ at all temperatures. The average photon number in the cavity is given by $\langle a^\dagger a\rangle = \alpha^2$, which clearly scales as N. We see, as well, that close to the transition α scales as $\alpha \propto (\lambda - \lambda_c)^\beta$ with $\beta = 1/2$, which indicates that the phase transition belongs to the mean field Ising universality class [5].

1.3 The first no-go theorem

Despite its theoretical interest, the phenomenon of photon condensation is yet to be achieved experimentally. This sparked an ongoing debate in the community that revolves around the validity of the approximations made to reach Dicke's model (4) from the full Hamiltonian (3). In the Dicke model, the coupling between the *electric dipole* and the cavity is generated by minimal coupling, which is, however, truncated at first order in A. The first no-go theorem arrived shortly after Hepp and Lieb's discovery, in 1975, and it pertained to the adequacy of neglecting the A^2 term [6]. Rzazewski *et al.* showed that the inclusion of the neglected term forbid the superradiant transition *via electric dipole coupling*, concluding that photon condensation is *an interesting artifact* (sic) of the Dicke model arising exclusively from the absence of the A^2 term, but without relevance in the description of real matter. We will not dwell reproducing their full argument, but instead give a brief and straightforward reasoning as to why the inclusion of the

A^2 term prevents the phase transition. Consider Hamiltonian (3) neglecting the Zeeman and spin-orbit coupling terms and in the long wave-length approximation, this yields

$$\mathcal{H} = \sum_i \frac{\left(\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}\right)^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c a^\dagger a. \tag{19}$$

To ignore the A^2 term one must develope the square in the first summand, leaving only the pure kinetic term of p^2 and the term from which electric coupling arises $\boldsymbol{pA}$. We have seen how this leads to the Dicke model in which photon condensation is present. Consider now that we keep the A^2 term and as such, the square of the first summand can be left as is. The partition function of this system is computed by simply integrating over both real and momentum space, as well as over the photonic degrees of freedom. In order to do so, let us consider the bosonic operators as c-numbers, using bounds analogous to the ones presented in Eq. 8 (See App. B.2 for a detailed proof in this instance), and write

$$\mathcal{H}(\alpha) = \sum_i \frac{\left(\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}\right)^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c |\alpha|^2. \tag{20}$$

Where $\boldsymbol{A} = 2\boldsymbol{A}_0 \operatorname{Re}(\alpha)$ now. The trace is invariant under unitary transformations, so a unitary transformation that displaces the momenta leaves the partition function unchanged. In essence the system behaves as if we had not introduced minimal coupling.

$$\begin{aligned} &Tr\left(\exp\left[-\beta\left(\sum_i \frac{\left(\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}\right)^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c|\alpha|^2\right)\right]\right) \equiv \\ &\equiv Tr\left(\exp\left[-\beta\left(\sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c|\alpha|^2\right)\right]\right) \end{aligned} \tag{21}$$

This implies that the restoration of gauge invariance effectively decouples light and matter, leaving a system incapable of experiencing a phase transition from light-matter interaction as is photon condensation.

1.4 Subsequent no-go and counter-no-go theorems

The first proposal to elude the A^2 term veered towards circuit QED, arguing that the TRK sum rule presented above is violated in systems of cooper pair boxes capacitively coupled to resonators [7]. Still, it was later argued that the sum rule applied in these systems as well [8]. Other proposals to bypass the A^2 term have studied graphene in the fractional quantum Hall regime [9], but they have been equally countered in subsequent publications [10]. These strategies rely on finding systems where the A^2 term is, supposedly, naturally absent, but they have always been countered by showing that the A^2 term was in fact present but neglected, or that it is *dynamically generated* by some mechanism.

More recently, the argument has shifted and it is now centered around gauge-related issues. All the original no-go theorems were proven in the Coulomb gauge (such as the one discussed in Sec. 1.3), in which Hamiltonian (3) is written. A Power-Zienau-Wolley (PZW) transformation eliminates the A^2 term in the dipole gauge, apparently resolving the long standing conflict [11]. This is however unsatisfactory, since physical predictions must be gauge-invariant, so finding a phase transition in a gauge that is prohibited in another is not a result that we can accept lightly. The most recent contribution to the topic (to the best of our knowledge), attempts to unify the discussion, claiming that the preceding no-go and counter no-go theorems are but two sides of the same coin [13]. According to the authors, the different predictions found in the two gauges can be explained by the fact that the physical fields associated to the photonic operators $a, a^\dagger$ are also gauge dependent. Though the idea is initially attractive, their argumentation is rather involved, and they mistakenly center the debate around the idea that previous predictions found a ferroelectric phase transition in the Coulomb gauge that presented as superradiant in the dipole gauge. Their paper reconciles these two manifestations of a phase transition, but in doing so, it misses the point. The phase transition that they describe and study arises solely from the inclusion of direct matter-matter interactions and it is not caused by the coupling to light, as a proper superradiant phase transition ought to be.

In Sec. 2, we settle this discussion by presenting a no-go theorem in the dipole gauge and unifying it with previous no-go theorems in the Coulomb gauge.

Another recent uncontested counter no-go theorem comes from the field of circuit QED [21]. The authors devise a particular superconducting circuit in which, they claim, superradiance occurs. In Sec. 2 we reply to their proposal, showing that our no-go theorem applies to their case as well.

As this document was being finalized, we learned about two newly published preprints that study the topic of photon condensation by considering spatially varying fields in the cavity and their interactions with 2D and 3D interacting electron systems [22, 23]. They support the existence of photon condensation in that context, but, in any case, they do not clash with our results.

1.5 A novel theoretical approach to the no-go theorem

In September of 2019 Andolina *et al.* published a no-go theorem in the Coulomb gauge for a very general matter Hamiltonian at $T = 0$, including electron-electron interactions and without applying the TLA [12]. The elegance and apparent definiteness of their proof caught our attention, and motivated our interest on the topic. Due to the relevance of their work, specially since we will later use and extend their formalism in ours, it seems appropriate to reproduce their proof here.

Consider an expansion of Hamiltonian (19) in which only Zeeman and spin-orbit coupling terms are ignored

$$\mathcal{H} = \mathcal{H}_m + \hbar\omega_c a^\dagger a - \sum_i \frac{e}{mc}\boldsymbol{p}_i \boldsymbol{A}_0 \left(a + a^\dagger\right) + \sum_i \frac{e^2 \boldsymbol{A}_0^2}{2mc^2}\left(a + a^\dagger\right)^2 \tag{22}$$

where $\mathcal{H}_m$ refers to the terms including only matter degrees of freedom

$$\mathcal{H}_m = \sum_i \frac{\boldsymbol{p}^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j). \tag{23}$$

In their work, the authors consider different effective masses m_i for the electrons, but this detail is not central to the proof so it is omitted here for simplicity. It is convenient to define two quantities that appear repeatedly through the proof:

$$\boldsymbol{j}_p \equiv \sum_i \frac{\boldsymbol{p}_i}{m}; \qquad \Delta \equiv \sum_i \frac{e^2 \boldsymbol{A}_0^2}{2mc^2}, \tag{24}$$

such that

$$\mathcal{H} = \mathcal{H}_m + \hbar\omega_c a^\dagger a - \frac{e}{c}\boldsymbol{j}_p \boldsymbol{A}_0 \left(a + a^\dagger\right) + \Delta \left(a + a^\dagger\right)^2. \tag{25}$$

A Bogoliubov transformation $-b = \cosh(x)a + \sinh(x)a^\dagger$, with $\cosh(x) = (\lambda + 1)/(2\sqrt{\lambda})$ and $\sinh(x) = (\lambda - 1)/(2\sqrt{\lambda})$ where $\lambda = \sqrt{1 + 4\Delta/(\hbar\omega_c)}$, allows us to eliminate the off-diagonal terms yielding

$$\mathcal{H} = \mathcal{H}_m + \hbar\omega_c \lambda b^\dagger b + \frac{e}{c}\boldsymbol{j}_p \boldsymbol{A}_0 \lambda^{-1/2} \left(b + b^\dagger\right). \tag{26}$$

We can divide the Hamiltonian (26) into matter and light terms, $\mathcal{H} = \mathcal{H}_m + \mathcal{H}_{ph} + \mathcal{H}_{int}$, which are extensive, i.e. they scale as $\sim N$. We can define intensive analogues that are well defined in the thermodynamic limit and prove that they commute in the thermodynamic limit (See App. D.1 for further details). Thus, for $N \to \infty$, matter and light become separable. The ground state is of the form $|\Psi\rangle = |\alpha\rangle \otimes |\psi\rangle$, where $|\alpha\rangle$ is a coherent state for the photons in the cavity to be determined by minimization of the energy, and $|\psi\rangle$ is the matter wave function. The energy of such a state is

$$E_\psi(\alpha) = \langle\psi|\mathcal{H}_m|\psi\rangle + \frac{e}{c}\langle\psi|\boldsymbol{j}_p|\psi\rangle \boldsymbol{A}_0 \frac{2\alpha}{\sqrt{\lambda}} + \hbar\omega_c\lambda\alpha^2. \tag{27}$$

$E_\psi(\alpha)$ needs to be minimized with respect to α and ψ. The former can be done analytically, yielding

$$\alpha = -\frac{1}{\hbar\omega_c\lambda^{3/2}}\frac{e}{c}\langle\psi|\boldsymbol{j}_p|\psi\rangle \boldsymbol{A}_0. \tag{28}$$

Substituting back into Eq. (27) provides an effective energy $E_\psi(\alpha) = \langle\psi|\mathcal{H}_m|\psi\rangle - \hbar\omega_c\lambda\alpha^2$, for which the ground state $|\psi\rangle$ is to be found while observing the restriction imposed by the minimization condition (28). This constitutes a constrained minimization problem that can be solved with the stiffness theorem (See App. E.1). Photon condensation will occur if the true minimum is a state with $\alpha \neq 0$, that is, if $E_\psi(\alpha) < E_{\psi_0}(0)$ where $|\psi_0\rangle$ is the true ground state of $\mathcal{H}_m$. This condition demands $\langle\psi|\mathcal{H}_m|\psi\rangle - \langle\psi_0|\mathcal{H}_m|\psi_0\rangle < \hbar\omega_c\lambda\alpha^2$, of which the left-hand side can expressed as a second order power of α using the stiffness theorem, obtaining

$$\langle\psi|\mathcal{H}_m|\psi\rangle - \langle\psi_0|\mathcal{H}_m|\psi_0\rangle = -\frac{1}{\chi}\frac{\alpha^2}{2}. \tag{29}$$

Where the susceptibility χ can be calculated to be (See App. F)

$$\chi = -\frac{2}{\hbar^2\omega_c^2\lambda^3}\frac{e^2}{c^2}\sum_{n\neq 0}\frac{|\langle\psi_n|\,\boldsymbol{j}_p\boldsymbol{A}_0\,|\psi_n\rangle|^2}{\epsilon_n - \epsilon_0}. \tag{30}$$

Accordingly, photon condensation requires

$$4\frac{e^2}{c^2}\sum_{n\neq 0}\frac{|\langle\psi_n|\,\boldsymbol{j}_p\boldsymbol{A}_0\,|\psi_n\rangle|^2}{\epsilon_n - \epsilon_0} > \hbar\omega_c + 4\Delta. \tag{31}$$

But the left hand side can be shown to be 4Δ through a TRK sum rule (See App. G). The resulting critical condition $4\Delta > \hbar\omega_c + 4\Delta$ is never satisfied, proving that photon condensation is indeed forbidden. Moreover, it is now clear that the ground state of the system is $|\psi_0\rangle \otimes |0\rangle$ regardless of the coupling. The strength of this proof relies on the fact that it avoids the typical shortcomings of no-go theorems in the Coulomb gauge. It does not rely on the TLA and it takes into consideration the Coulomb interaction between electrons. It also presents a novel way of defining the critical condition in terms of the susceptibility, which we will extend in Sec. 3.2.

2 Settling the debate

Thus far, we have shown that the debate does not revolve around whether or not the Dicke model presents superradiance; it does. We have presented proof in the preceding section. The argument lies in whether the Dicke model is an appropriate many body simplification of the Pauli equation or if, on the contrary, new terms and features must be included that, upon consideration, prohibit photon condensation. The inclusion of the A^2 term restores gauge invariance and, quite simply, prohibits superradiance in the Coulomb gauge. This should be sufficient to put to rest the argument, as *the physics of the model cannot depend on the choice of gauge*, yet it has been dragged on for years on the basis that the dipole gauge showed signs of allowing the phase transition. More recently, the debate has become even more convoluted by considering matter systems that presented phase transitions on their own, confusing them with the superradiant phase transition. In this section we use the bounds proven for Z (8), in the thermodynamic limit, to present a no-go theorem in the dipole gauge at finite T. We then unify this no-go theorem with the already existing no-go theorems in the Coulomb gauge, settling the debate.

Let us apply a PZW transformation to switch to the dipole gauge. Consider a unitary transformation of the form

$$U = \exp\left[-\frac{i}{c\hbar}F\right]; \qquad \text{with} \qquad F = -\sum_i e\boldsymbol{r}_i\boldsymbol{A} = \boldsymbol{d}\boldsymbol{A}. \tag{32}$$

We have introduced $\boldsymbol{d}$ the total electric dipole operator. The change of gauge amounts to applying the PZW transformation onto Hamiltonian (22): $\mathcal{H}' = U^\dagger \mathcal{H} U$, yielding (See App H for a full derivation)

$$\mathcal{H}' = \mathcal{H}'_m + \hbar\omega_c a^\dagger a - i\frac{\omega_c}{c}\boldsymbol{d}\boldsymbol{A}_0\left(a^\dagger - a\right), \tag{33}$$

where

$$\mathcal{H}'_m = \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \frac{\omega_c}{c^2\hbar}(\boldsymbol{d}\boldsymbol{A}_0)^2. \tag{34}$$

Taking the bosonic operators as c-numbers yields (See App. B.3 for proof that this switch is also valid in the dipole gauge)

$$\mathcal{H}'(\alpha) = \mathcal{H}'_m + \hbar\omega_c|\alpha|^2 - 2\frac{\omega_c}{c}\boldsymbol{d}\boldsymbol{A}_0\,\mathrm{Im}(\alpha). \tag{35}$$

It is convenient, now, to obtain an effective spin Hamiltonian were the light degrees of freedom have been traced out. From Eq. (12) we can define the effective spin Hamiltonian

$$\bar{Z} = Tr_m\left(\frac{1}{\pi}\int d^2\alpha\; e^{-\beta\mathcal{H}'(\alpha)}\right) = Tr_m\left(e^{-\beta H_{\text{eff}}}\right). \tag{36}$$

Thus

$$\frac{1}{\pi}\int d^2\alpha\; e^{-\beta\mathcal{H}'(\alpha)} =$$
$$\frac{1}{\pi}\int d\,\mathrm{Re}(\alpha)\; e^{-\beta\hbar\omega_c\,\mathrm{Re}(\alpha)^2}\int d\,\mathrm{Im}(\alpha)\;\exp\left[-\beta\left(\mathcal{H}'_m + \hbar\omega_c\,\mathrm{Im}(\alpha)^2 - 2\frac{\omega_c}{c}\boldsymbol{d}\boldsymbol{A}_0\,\mathrm{Im}(\alpha)\right)\right]. \tag{37}$$

These are two Gaussian integrals that can be easily computed. The integral over $\mathrm{Re}(\alpha)$ yields only a constant factor, it is the integral over $\mathrm{Im}(\alpha)$ that yields a modification to $\mathcal{H}'_m$ such that

$$\mathcal{H}_{\text{eff}} = \mathcal{H}'_m - \frac{\omega_c}{c^2\hbar}(\boldsymbol{d}\boldsymbol{A}_0)^2 = \mathcal{H}_m. \tag{38}$$

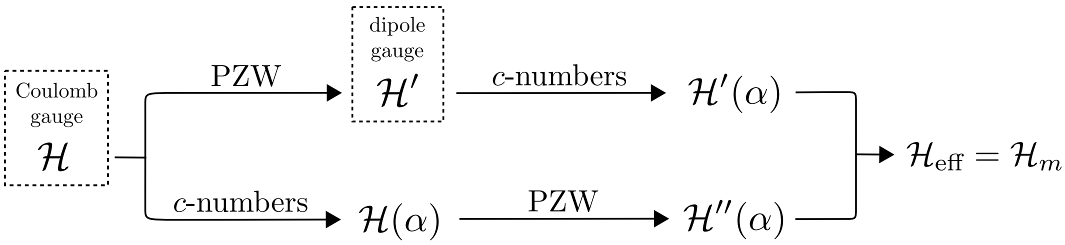

Fig. 2: Schematic diagram of the different routes to reach $\mathcal{H}_{\text{eff}} = \mathcal{H}_m$ as the final result of a no-go theorem.

We have used the fact that $[\mathcal{H}'_m/N, \mathcal{H}_{int}/N] \to 0$ in the thermodynamic limit (See App. D.2) in order to extract the exponential of $\mathcal{H}'_m$ from the imaginary integral before integration and again when putting together $\mathcal{H}_{\text{eff}}$. The effective Hamiltonian is simply $\mathcal{H}_m$, proving that in the thermodynamic limit the coupling to the cavity does not play any role in the ground state for the matter system and no cavity-coupling induced transition occurs in the dipole gauge. This is a main result of our work. Previously, authors were able to prove counter no-go theorems in the dipole gauge, i.e. that photon condensation was possible in this gauge, because they ignored the A^2 term in the dipole gauge [11]. The authors neglected it when performing the TLA on the matter Hamiltonian because in this gauge it no longer contains photonic operators and it is grouped with the matter terms [Cf. Eq. (35)]. As we just saw, the inclusion of this term precisely cancels the effective spin-spin interaction mediated by the cavity, and thus *its neglect is the sole origin of the superradiant phase transition* in the dipole gauge.

To bring closure to this topic, let us show how to unify the no-go theorems in the Coulomb and dipole gauges. Consider the Hamiltonian in the Coulomb gauge after substituting the bosonic operators for c-numbers (20). We can apply the PZW transformation now, which at this point is simply a displacement operator on the momentum $U(\boldsymbol{p}_i - e/c\boldsymbol{A})U^\dagger \to \boldsymbol{p}_i$. The resulting Hamiltonian $\mathcal{H}''(\alpha)$ has no light-matter interaction

$$\mathcal{H}''(\alpha) = \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c|\alpha|^2 = \mathcal{H}_m + \hbar\omega_c|\alpha|^2, \tag{39}$$

so the effective Hamiltonian [1] in this case is simply $\mathcal{H}_{\text{eff}} = \mathcal{H}_m$, proving again that no superradiant phase transition is possible. Even though the PZW transformation and the substitution of the bosonic operators by c-numbers are not commuting operations, in the sense that $\mathcal{H}''(\alpha) \neq \mathcal{H}'(\alpha)$, when we compute the effective Hamiltonian in both cases, the result is the same. This is illustrated in Fig. 2 for clarification. The final issue to discuss is the relation between light and matter operators in the different gauges. Consider a in the Coulomb gauge, and let us compute its average value after the switch to c-numbers and the subsequent *PZW* transformation to displace the momenta

$$\langle a \rangle = \frac{1}{\bar{Z}} Tr_m \left(\frac{1}{\pi} \int d^2\alpha \; \alpha \; e^{-\beta(\mathcal{H}_m + \hbar\omega_c|\alpha|^2)} \right) = 0. \tag{40}$$

Thus $\langle a \rangle$ is disconnected from matter operators. Regardless whether or not the bare matter system experiences a phase transition, e.g. a ferroelectric one, that yields a non zero value

[1] The effective Hamiltonian is defined analogously to the dipole gauge case $\bar{Z} = Tr_m \left(\frac{1}{\pi}\int d^2\alpha \; \exp\left[-\beta\mathcal{H}''(\alpha)\right]\right) = Tr_m \left(\exp\left[-\beta H_{\text{eff}}\right]\right)$

of $\langle \boldsymbol{d}\boldsymbol{A}_0 \rangle_m$, this will not translate into a finite value of $\langle \alpha \rangle$. Expectation values are gauge invariant, so this result is also true in the dipole gauge. Nevertheless, consider now the dipole gauge, in this case, the PZW transformation is performed before the c-number substitution, thus $a \to a - \frac{i}{c\hbar} \boldsymbol{d}\boldsymbol{A}_0 \equiv a'$. If, however, we insist on computing $\langle a \rangle'$ as an order parameter, instead of the gauge invariant $\langle a' \rangle'$, we obtain

$$\begin{aligned} \langle \alpha \rangle' &\propto \frac{1}{\bar{Z}} Tr_m \left(\frac{1}{\pi} \int d\,\mathrm{Im}(\alpha)\ \mathrm{Im}(\alpha)\ \exp\left[-\beta \left(\mathcal{H}'_m + \hbar\omega_c\, \mathrm{Im}(\alpha)^2 - 2\frac{\omega_c}{c} \boldsymbol{d}\boldsymbol{A}_0\, \mathrm{Im}(\alpha) \right) \right] \right) \\ &= \frac{1}{\bar{Z}} Tr_m \left(\frac{i}{c\hbar} \boldsymbol{d}\boldsymbol{A}_0 e^{-\beta \mathcal{H}_{\text{eff}}} \right) = \frac{i}{c\hbar} \langle \boldsymbol{d}\boldsymbol{A}_0 \rangle_m \end{aligned} \tag{41}$$

We see now that despite being effectively disentangled, the expectation values of light and matter operators are related. If the bare matter model presents a phase transition that makes $\langle \boldsymbol{d}\boldsymbol{A}_0 \rangle_m \neq 0$ this translates into a non-zero value of $\langle \alpha \rangle'$. Of course, this just an artifact arising from the fact that have not dealt with the transformed bosonic operator a' whose definition tells us that its expectation value is $\langle \alpha' \rangle' = \langle \alpha \rangle' - \frac{i}{c\hbar} \langle \boldsymbol{d}\boldsymbol{A}_0 \rangle_m = 0$. In light of these results, we must conclude that a finite value of $\langle \alpha \rangle'$ is not a proper indicator of the superradiant phase transition, it is only if this finite value is *caused by light-matter coupling* and not by a choice of gauge, that the phase transition is superradiant. Furthermore, this result supersedes the convoluted discussion in Ref. [13]. We present their results in a more general and straightforward fashion, and we point out that they were mistakenly interpreting a ferroelectric phase transition as a superradiant one.

In App. J we show how to map the Hamiltonian of the circuit QED model presented in Ref. [21] to a Hamiltonian in "minimal coupling form" such as Pauli's equation, and from there we prove that the no-go theorem we just presented applies to it as well. Because the theory of circuit QED requires introducing a new notation, breaking the tonic of the present document, we reserve this discussion for App. J, but the reader should take this as another main result of our work, and is encouraged to find all the details in the appendix.

3 Magnetic cavity QED

3.1 Zeeman coupling to bypass previous obstacles

It seems, at this point, that the only way to escape the no-go theorems presented in the previous section is to include more terms of the Pauli equation into our microscopic model of matter coupled to a cavity, the only two terms left being the Zeeman coupling and the spin-orbit coupling. We decided to consider the former as a first approximation to the issue. Spin-orbit coupling scales independently so it is expected that it can be neglected in most cases. This introduces the magnetic field into the picture, leading to what we have termed *magnetic cavity QED*. We view magnetic cavity QED as a way of avoiding all obstacles previously present when considering only the electric coupling between atoms and cavity. If we are able to ignore the electric coupling, which we are in the case of molecules or atoms with zero electric dipole moment, we arrive at a Hamiltonian with three key properties: (a) It is gauge invariant, as we will prove shortly; (b) in the appropriate cases, i.e. in magnetic molecules with free spin 1/2 electrons, the TLA is exact; (c) The A^2 term is not present, allowing us to reach the Dicke model, or, in some cases, a generalization of it.

Let us show how Hamiltonian (3) can lead to the Dicke model with the inclusion of the Zeeman coupling. We will rewrite it here keeping only the terms we are interested in and assuming a uniform cavity field

$$\begin{aligned}\mathcal{H} =& \sum_i \frac{\boldsymbol{p}^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c a^\dagger a - \sum_i \frac{e}{mc}\boldsymbol{p}_i \boldsymbol{A}_0 (a + a^\dagger) \\ &+ \sum_i \frac{e^2 \boldsymbol{A}_0^2}{2mc^2}(a + a^\dagger)^2 - \sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}_i \boldsymbol{B}_0 (a + a^\dagger).\end{aligned} \tag{42}$$

In order to make explicit its dependence on the electric dipoles, let us apply a PZW transformation to switch to the dipole gauge. Clearly if $\boldsymbol{d} = 0$ then $U = \mathbb{I}$: the transformation becomes the identity in the case of molecules with no electric dipole, but it is still illustrative to apply it. The Zeeman term is invariant under the PZW so recycling Eq. 33 we obtain

$$\begin{aligned}\mathcal{H}' =& \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c a^\dagger a - i\frac{\omega_c}{c}\boldsymbol{d}\boldsymbol{A}_0 \left(a^\dagger - a\right) \\ &+ \frac{\omega_c}{c^2\hbar}(\boldsymbol{d}\boldsymbol{A}_0)^2 - \sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}\boldsymbol{B}_0 \left(a + a^\dagger\right).\end{aligned} \tag{43}$$

We have made explicit the dependence of the problem on the electric dipole. Thus, for scenarios with zero electric dipole, which are feasible in architectures of molecules with free radicals or artificial atoms [24], the Hamiltonian, which takes the same expression both in the Coulomb and dipole gauges ($\mathcal{H}' = \mathcal{H}$), is

$$\mathcal{H}' = \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) + \hbar\omega_c a^\dagger a - \sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}\boldsymbol{B}_0 \left(a + a^\dagger\right). \tag{44}$$

At this point, the coupling between the electrons and the cavity comes exclusively from the Zeeman term. The matter part of the Hamiltonian will be determined phenomenologically, depending on the actual experimental setup, but we can make certain assumptions about its structure, for instance, that it will depend only on the spin angular momentum of the electrons

$$\mathcal{H}_m = \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_i V(\boldsymbol{r}_i) + \frac{1}{2}\sum_{i\neq j} v(\boldsymbol{r}_i - \boldsymbol{r}_j) \equiv \frac{\hbar\omega_z}{2}\sum_i \sigma_i^z + \mathcal{H}_c = \mathcal{H}_S. \tag{45}$$

Here $\mathcal{H}_S$ is the effective spin Hamiltonian. Without loss of generality, we can assume that it will be formed by a term corresponding to the Zeeman splitting of electronic energy levels and a term $\mathcal{H}_c$ corresponding to the effective coupling between spins. This coupling can be of Ising, Heisenberg or any other nature, and even non-existent in an idealized situation. We will later explore the multiple possibilities that it offers, but for now let us focus on the simpler case $\mathcal{H}_c = 0$. In the case of free radicals or artificial atoms, the Zeeman splitting is induced by the application of a classical, i.e. non-quantized, external field. For that reason ω_z is in general a tunable parameter. Having assumed that the cavity field is uniform, a condition which we will relax at some point, we can also assume that the magnetic field points in the x direction. Again, this can be chosen so in the experimental setup. As illustrated in Fig. 3, the cavity field is fixed by the geometry of the cavity but the external magnetic field $\boldsymbol{B}_{ext}$ responsible for the Zeeman splitting can be chosen to point in any direction, essentially setting the reference

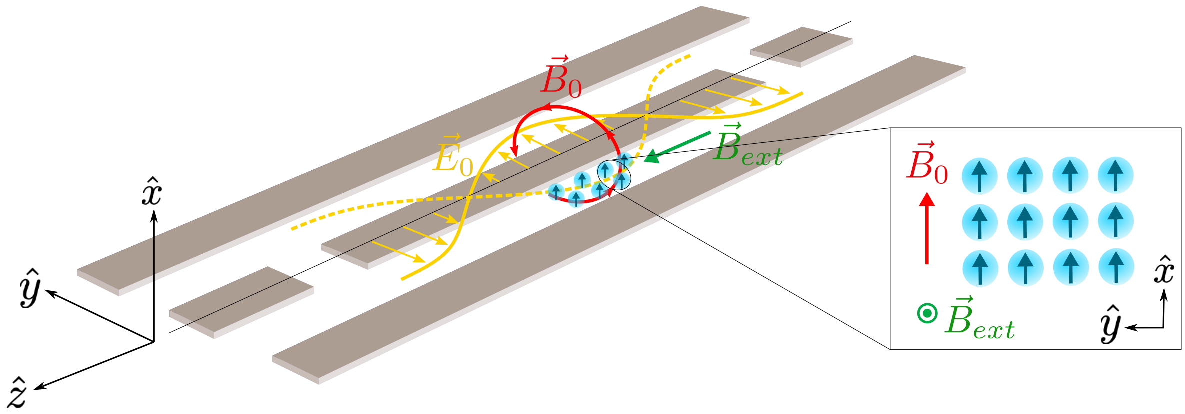

Fig. 3: Schematic depiction of a superconducting cavity with an ensemble of spins coupled to it. $\vec{E}_0$ and $\vec{B}_0$ are the cavity's electric and magnetic fields, respectively. $\vec{B}_{ext}$ is the external field used to induce the Zeeman splitting on the spin energy levels.

frame. Furthermore, we can relax the assumption that B_0 is uniform. Under such conditions, the coupling term becomes

$$-\sum_i \frac{e\hbar}{2mc}\boldsymbol{\sigma}\boldsymbol{B}_0(\boldsymbol{r}_i)\left(a+a^\dagger\right) = \sum_i \frac{e\hbar}{2mc}\sigma_i^x|B_{rms}(\boldsymbol{r}_i)|\left(a+a^\dagger\right). \tag{46}$$

Where $B^2_{rms}(\boldsymbol{r}_i) = \langle 0|\, B_0^2(\boldsymbol{r}_i)\,|0\rangle$ is the root mean square of the cavity's magnetic field vacuum fluctuations. Notice that this is precisely the Hamiltonian of the Dicke model with inhomogeneous coupling (4) with

$$\frac{\lambda_i}{\sqrt{N}} = \frac{e}{2mc}\sigma_i^x|B_{rms}(\boldsymbol{r}_i)|. \tag{47}$$

In experimental setups, λ_i must be calculated numerically. In the case of homogeneous coupling, we obtain plain Dicke model, and we can give an estimation of the value of $|B_{rms}|$ and thus λ using the Virial theorem, i.e. $E_{EM} = \frac{E_E}{2} + \frac{E_B}{2}$. The energy corresponding to vacuum fluctuations is $\frac{1}{2}\hbar\omega_c$, and we have

$$\frac{1}{2}\int dV \frac{\langle 0|\, B_0^2\,|0\rangle}{\mu_0} = \frac{1}{2}\int dV \frac{B^2_{rms}}{\mu_0} = \frac{1}{4}\hbar\omega_c. \tag{48}$$

This yields $|B_{rms}| = \sqrt{\frac{\hbar\omega_c\mu_0}{2V}}$, with V the volume of the cavity. Then,

$$\lambda = \frac{e}{2mc}\sqrt{\frac{\hbar\omega_c\mu_0}{2}\rho}. \tag{49}$$

With $\rho = N/V$ being the density of electrons in the cavity. It is convenient to express λ^2 in simpler terms as

$$\lambda^2 = 4\frac{\mu_B^2\mu_0}{2\hbar}\rho\omega_c = \eta\rho\omega_c, \tag{50}$$

where μ_B is Bohr's magneton and $\eta = 5\cdot 10^{-19}\ m^3 s^{-1}$. Comparing this with the electric dipole coupling obtained in the original formulation of the Dicke model (5), we find

$$\frac{\lambda_e}{\lambda_m} \sim \frac{mdc}{e\hbar} \sim \frac{ma_0c}{\hbar} \sim \frac{1}{\alpha} \approx 137. \tag{51}$$

With α the fine structure constant in this instance. As expected, the magnetic coupling is weaker than the electric one, but as Fig. 4 shows, the phase transition still occurs at experimentally

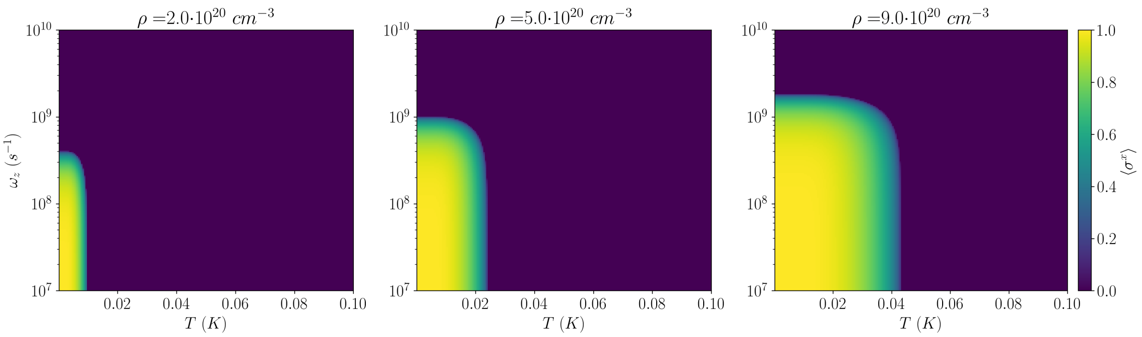

Fig. 4: Phase diagram for the Dicke model in terms of ρ, T and ω_z. The colormap shows the transverse magnetization per spin $\langle\sigma^x\rangle$ at equilibrium. A finite value is indicative of the superadiant phase, where $\langle\sigma^x\rangle \propto \alpha \neq 0$

accessible values of T and ω_z. If we recover the critical condition for the Dicke model (17) and substitute the new found value of λ^2 we obtain the critical condition $4\eta\rho = \omega_z \coth\left(\beta\frac{\omega_z}{2}\right)$, which depends only on the electron density ρ and ω_z, not on ω_c. , Experiments have been performed already with similar setups, and they report densities in the range of $2 - 9 \cdot 10^{20}$ cm^{-3} [25]. In Fig. 4 we show the phase diagram of the system for several values of ρ lying within this range. As we can see, a higher concentration of electrons facilitates the phase transition. We also observe that the values of the critical temperature and critical ω_z are well within current experimental capabilities.

3.2 Beyond the Dicke model

We have just seen that a larger density of spins facilitates the phase transition in the basic Dicke model obtained by considering uniform Zeeman coupling with non-interacting spins. Consequently, in an experimental search of the phase transition, we would be inclined to use the highest density achievable. At such high concentrations of spins, two of our previous assumptions will be broken: if the spins are closely packed, at some point the interaction among them will become non-negligible forcing us to consider $\mathcal{H}_c \neq 0$; moreover, since the density ρ is defined as the number of spins divided by the volume of the cavity, maximizing it would require depositing spins in the whole cavity volume, which breaks the approximation of uniform field. Hence, we must consider a generalized version of the previous model, that includes direct spin-spin interactions and spin-dependent coupling to the magnetic field. That is, a Hamiltonian of the form

$$\mathcal{H} = \omega_c a^\dagger a + \mathcal{H}_S + \frac{1}{\sqrt{N}}(a + a^\dagger)\sum_j \lambda_j \sigma_j^x. \tag{52}$$

We found that a generalization of Andolina and colleagues' approach is convenient for dealing with general Hamiltonians of this sort. Let us first consider the zero temperature case, and then advance to finite temperature.

3.2.1 Zero temperature $T = 0$

In the thermodynamic limit, $N \to \infty$, it can be shown that $[\mathcal{H}_{ph}, \mathcal{H}_{int}] = [\mathcal{H}_S, \mathcal{H}_{int}] = 0$ (See App. D.3), thus light and spin degrees of freedom are effectively decoupled and the states of the system are separable. In such scenario a mean-field treatment becomes exact, and we can propose for the photons in the cavity a coherent state $a\,|\alpha\rangle = \alpha\,|\alpha\rangle$. Taking $\alpha \in \mathbb{R}$ and a spin state $|\psi\rangle$, we find

$$E(\alpha) = \omega_c \alpha^2 + \langle\psi|\mathcal{H}_S|\psi\rangle + \frac{2\alpha}{\sqrt{N}}\,\langle\psi|\sum_j \lambda_j \sigma_j^x|\psi\rangle\,. \tag{53}$$

Minimizing with respect to α yields

$$\langle\psi|\sum_j \lambda_j \sigma_j^x|\psi\rangle = -\sqrt{N}\omega_c\alpha, \tag{54}$$

which constitutes a constrain in the search of the ground state of $\mathcal{H}_S$. Upon substituting back, Eq. (54) gives the minimum energy for a state with non-zero α

$$E_\psi(\alpha) = \langle\psi|\mathcal{H}_S|\psi\rangle - \omega_c\alpha^2. \tag{55}$$

For the state $|\Psi\rangle = |\psi\rangle \otimes |\alpha\rangle$ with $\alpha \neq 0$ to be the true minimum requires $|\psi\rangle$ to be the least energetic state obeying the constrain (54) (we term it the *constrained ground state*) and, of course, $E_\psi(\alpha) \leq E_{\psi_0}(0)$, i.e.

$$\langle\psi|\mathcal{H}_S|\psi\rangle - \langle\psi_0|\mathcal{H}_S|\psi_0\rangle \leq \omega_c\alpha^2. \tag{56}$$

Where $|\psi_0\rangle$ is the unconstrained ground state of $\mathcal{H}_S$. The stiffness theorem allows us to expand $\langle\psi|\mathcal{H}_S|\psi\rangle - \langle\psi_0|\mathcal{H}_S|\psi_0\rangle$ in powers of α (see App. E.1). Provided $\langle\psi_0|\sum_j \lambda_j\sigma_j^x|\psi_0\rangle = 0$, which will be true at least for all parity-conserving spin Hamiltonians, we find

$$\langle\psi|\mathcal{H}_S|\psi\rangle - \langle\psi_0|\mathcal{H}_S|\psi_0\rangle = -\frac{\alpha^2}{2\chi}, \tag{57}$$

with χ being the susceptibility (see App. F)

$$\chi = -\frac{2}{N\omega_c}\sum_{m\neq 0}\frac{|\,\langle\psi_m|\sum_j \lambda_j\sigma_j^x\,|\psi_0\rangle\,|^2}{\epsilon_m - \epsilon_0}. \tag{58}$$

$|\psi_m\rangle$ and ϵ_m are respectively the eigenstates and eigenenergies of $\mathcal{H}_S$ such that $\mathcal{H}_S\,|\psi_m\rangle = \epsilon_m\,|\psi_m\rangle$. From Eqs. (56)-(58) we obtain the condition for the superradiant phase transition of a general spin Hamiltonian $\mathcal{H}_S$

$$\frac{\omega_c}{4} \leq \frac{1}{N}\sum_{m\neq 0}\frac{|\,\langle\psi_m|\sum_j \lambda_j\sigma_j^x\,|\psi_0\rangle\,|^2}{\epsilon_m - \epsilon_0}. \tag{59}$$

This inequality might resolved by exactly solving the spin model or, alternatively, by any exact-diagonalization technique able to provide all eigenstates that have non-negligible contributions to the sum in the right hand side. As a sanity check, an example of a solvable system is the Dicke model, for which the spin Hamiltonian is simply

$$\mathcal{H}_S = \frac{\omega_z}{2}\sum_j \sigma_j^z. \tag{60}$$

The ground state is trivially $|\psi_0\rangle = |00\ldots 0\rangle \equiv |0\rangle$ and it only connects with single-excitation states in the sum, i.e. $|0\ldots 1_m \ldots 0\rangle \equiv |1_m\rangle$. Where $|0\rangle$, $|1\rangle$ are the eigenstates of σ^z with eigenvalues -1, $+1$, respectively. We thus find

$$\frac{1}{N}\sum_m \frac{|\,\langle 1_m|\sum_j \lambda_j\sigma_j^x\,|0\rangle\,|^2}{\omega_z} = \frac{1}{N}\sum_m\frac{|\sum_j \lambda_j\delta_{j,m}|^2}{\epsilon_m - \epsilon_0} = \frac{1}{N}\sum_m \frac{\lambda_m^2}{\omega_z} = \frac{\bar{\lambda}^2}{\omega_z}. \tag{61}$$

Plugging this into Eq. 59 yields

$$\bar{\lambda} \geq \frac{1}{2}\sqrt{\omega_c \omega_z}, \tag{62}$$

which is the well known critical condition for the Dicke model. Notice that it matches the critical condition obtained previously (17), where the uniform coupling considered originally is now replaced by a root-mean-square coupling defined from the spin-dependent couplings as $\bar{\lambda}^2 = N^{-1}\sum_m \lambda_m^2$. Let us show that, just as in the case of homogeneous coupling, this root-mean-square coupling can also be related to the density of spins. In the continuum limit, and using again the Virial theorem

$$\bar{\lambda}^2 = \frac{1}{V}\int dV \lambda(\boldsymbol{r}) = \rho\left(\frac{e}{2mc}\right)^2 \int dV B_{rms}^2(\boldsymbol{r}) = \rho\left(\frac{e}{2mc}\right)^2 \frac{\hbar\omega_c \mu_0}{2}. \tag{63}$$

Hence, $\bar{\lambda}^2 = \eta\rho\omega_c$.

3.2.2 Finite temperature $T \geq 0$

Firstly, let us assume that minimizing the energy is either sufficiently or completely equivalent to minimizing the free energy at low temperatures (See App. I). In that case, after considering a coherent state for the cavity, we can write an α-dependent Hamiltonian

$$\mathcal{H}(\alpha) = \omega_c \alpha^2 + \mathcal{H}_S + \frac{2\alpha}{\sqrt{N}}\sum_j \lambda_j \sigma^x. \tag{64}$$

If we consider that our thermal spin ensemble is captured by the density matrix ρ, the energy of the system is

$$E(\alpha) = \omega_c \alpha^2 + Tr(\mathcal{H}_S \rho) + \frac{2\alpha}{\sqrt{N}} Tr(\sum_j \lambda_j \sigma_j^x \rho), \tag{65}$$

which we can minimize with respect to α to yield a constrained minimum condition for the spin system

$$Tr(\sum_j \lambda_j \sigma_j^x \rho) = \langle \sum_j \lambda_j \sigma_j^x \rangle = -\sqrt{N}\omega_c \alpha. \tag{66}$$

Reintroducing it in our expression for the energy gives

$$E_\rho(\alpha) = Tr(\mathcal{H}_S \rho) - \omega_c \alpha^2. \tag{67}$$

With that, the critical condition becomes $E_\rho(\alpha) \leq E_{\rho_0}(0)$, which amounts to

$$Tr(\mathcal{H}_S \rho) - Tr(\mathcal{H}_S \rho_0) \leq \omega_c \alpha^2. \tag{68}$$

We can now make use of the extended stiffness theorem (See App. E.2) to write the left hand side of Eq. (68) in powers of α, to obtain a critical condition analogous to the zero temperature case, but with a susceptibility that is now temperature dependent

$$-\frac{1}{2}\frac{\alpha^2}{\chi(T)} \leq \omega_c \alpha^2 \rightarrow -\chi(T) \geq \frac{1}{2\omega_c}. \tag{69}$$

Considering the temperature dependent susceptibility (See App. F) we obtain the critical condition

$$\frac{\omega_c}{2} \leq \frac{1}{N}\frac{\sum_{m,n} e^{-\beta\epsilon_m} |\langle\psi_m| \sum_j \lambda_j \sigma_j^x |\psi_n\rangle|^2 \frac{e^{\beta\Delta_{mn}}-1}{\Delta_{mn}}}{\sum_m e^{-\beta\epsilon_m}}. \tag{70}$$

Where $|\psi_m\rangle$ and ϵ_m are respectively the eigenstates and eigenenergies of $\mathcal{H}_S$ and $\Delta_{mn} = \epsilon_m - \epsilon_n$. We will again check the validity of this result by comparing against the Dicke model for which we have an exact solution. For simplicity, we consider the standard Dicke model with homogeneous coupling $\lambda_j = \lambda$. In the Dicke model, the states are distributed in subspaces of fixed energy, each subspace is formed by vectors with a given amount of flipped spins with respect to the ground state. In this notation we denote $H(S)$ to the subspace with S flipped spins. Accordingly, all states in $H(S)$ have energy (above the ground state) $\epsilon(S) = \omega_z S$. Note that the number of states in a particular subspace $H(S)$ is $\Omega(S) = \binom{N}{S}$. Considering this, we can already rewrite

$$\sum_m e^{-\beta\epsilon_m} = \sum_S \Omega(S) e^{-\beta\omega_z S} = \sum_S \binom{N}{S} e^{-\beta\omega_z S} = \left(e^{-\beta\frac{\omega_z}{2}} 2\cosh\left(\beta\frac{\omega_z}{2}\right)\right)^N, \tag{71}$$

A result which is also readily obtained by factorizing the partition function. Focusing now on the upper sum of Eq. (70), notice that $\sum_j \sigma_j^x$ only connects states that belong to subspaces differing in one flipped state. With this in mind the sum $\sum_{m,n}$ becomes $2\sum_{m>n}$, which we can rewrite as

$$\sum_{m>n} \to \sum_S^{N-1} \sum_{\psi_m\in H(S+1)} \sum_{\psi_n\in H(S)}. \tag{72}$$

Furthermore, we have

$$\frac{e^{-\beta\epsilon_n} - e^{-\beta\epsilon_m}}{\epsilon_m - \epsilon_n} \to \frac{e^{-\beta\omega_z S} - e^{-\beta\omega_z(S+1)}}{\omega_z}, \tag{73}$$

which can be expressed as

$$e^{\beta\omega_z S} e^{-\beta\frac{\omega_z}{2}} \frac{2\sinh\left(\beta\frac{\omega_z}{2}\right)}{\omega_z}. \tag{74}$$

So the sum has become

$$2e^{-\beta\frac{\omega_z}{2}} \frac{2\sinh\left(\beta\frac{\omega_z}{2}\right)}{\omega_z} \sum_S^{N-1} e^{\beta\omega_z S} \sum_{\psi_m\in H(S+1)} \sum_{\psi_n\in H(S)} |\langle\psi_m| \sum_j \sigma_j^x |\psi_n\rangle|^2. \tag{75}$$

After some counting, one finds

$$\sum_{\psi_m\in H(S+1)} \sum_{\psi_n\in H(S)} |\langle\psi_m| \sum_j \sigma_j^x |\psi_n\rangle|^2 = \Omega(S+1)(S+1) = \binom{N}{S+1}(S+1). \tag{76}$$

So we are left to calculate

$$\sum_S^{N-1} e^{\beta\omega_z S} \binom{N}{S+1}(S+1) = N\left(e^{-\beta\frac{\omega_z}{2}} 2\cosh\left(\beta\frac{\omega_z}{2}\right)\right)^{N-1}. \tag{77}$$

Putting everything together, we find that the right hand side of Eq. (70) amounts to

$$\frac{2\lambda^2}{\omega_z} \tanh\left(\beta\frac{\omega_z}{2}\right). \tag{78}$$

Which in turn provides the critical condition for the Dicke model

$$\lambda \geq \frac{1}{2}\sqrt{\omega_z\omega_c \coth\left(\beta\frac{\omega_z}{2}\right)}, \tag{79}$$

which matches the one found through the original treatment of the Dicke model (17), thus validating our theory.

The *power of this formalism* lies in the fact that it allows us to handle any spin Hamiltonian $\mathcal{H}$ with a unique treatment. The susceptibility can be computed either analytically, in some simple cases where an exact solution of the spin model is known, e.g. the Dicke model; or numerically, in cases when an exact analytical solution is too cumbersome or simply unattainable. In addition, a numerical solution with this method is advantageous with respect to a numerical solution of the complete light-matter Hamiltonian. Considering that we have eliminated the light degrees of freedom, we only face an exact diagonalization of the spins Hamiltonian, which has lower dimensionality than the whole, and is thus more tractable. The disadvantage is that with this method we are only able to compute the phase boundary, and not observables such as $\langle\sigma^x\rangle$. In order to tackle this shortcomings, in the next subsection we present an alternative solution, where we calculate an effective Hamiltonian for the spins, from which observables can be computed.

3.3 Effects of light-matter coupling on the effective spin model: Ising coupling

In order to study particular spin models and how their properties are affected by the coupling to light, it is convenient to obtain an effective spin Hamiltonian were the light degrees of freedom have been traced out. Analogously to previous examples, we can define the effective spin Hamiltonian in this case (See App. B.3 for a proof on the bounds for Z in this case)

$$\bar{Z} = \frac{1}{\pi}\int d^2\alpha \; Tr_S\left(e^{-\beta\mathcal{H}(\alpha)}\right) = Tr_S\left(e^{-\beta H_{\text{eff}}}\right). \tag{80}$$

Where, for a general spin Hamiltonian, we define $\mathcal{H}(\alpha)$ as

$$\mathcal{H}(\alpha) = \mathcal{H}_S + \omega_c|\alpha|^2 + \frac{\lambda}{\sqrt{N}}\sum_j \sigma_j^x(\alpha+\alpha^*). \tag{81}$$

Thus

$$\frac{1}{\pi}\int d^2\alpha \; e^{-\beta\mathcal{H}(\alpha)} \propto \frac{1}{\pi}\int d\,\mathrm{Re}(\alpha)\; e^{-\beta\left(\mathcal{H}_S+\omega_c\,\mathrm{Re}(\alpha)^2+\frac{\lambda}{\sqrt{N}}\sum_j\sigma_j^x\,\mathrm{Re}(\alpha)\right)}. \tag{82}$$

Which yields

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_S - \frac{\lambda^2}{\omega_c N}\left(\sum_j \sigma_j^x\right)^2. \tag{83}$$

We have used the fact that $[\mathcal{H}_S/N, \mathcal{H}_{int}/N] \to 0$ in the thermodynamic limit (See App. D.3 for proof) in order to extract the exponential of $\mathcal{H}_S$ for integration and then to put together $\mathcal{H}_{\text{eff}}$. It is important to note that Eq. (83) is an exact result, equivalent to the one obtained with the extension of Andolina and colleagues' method in Eq. (70). We can now study the spectrum of $\mathcal{H}_{\text{eff}}$ with different techniques, which can include perturbation theory, mean field theory or even an exact analytical solution, depending on the nature of $\mathcal{H}_S$. In particular, we find mean field theory to be a convenient tool to obtain quick qualitative predictions on the behavior of the system. A mean field approximation of this Hamiltonian reads

$$\mathcal{H}_{\text{eff}}^{\text{MF}} = \mathcal{H}_S^{\text{MF}} - \frac{2\lambda^2}{\omega_c}m_x\sum_j\sigma_j^x + \frac{\lambda^2 N}{\omega_c}m_x^2, \tag{84}$$

where m_x is the per-spin transverse magnetization $m_x = \langle\sigma^x\rangle = \sum_j \sigma_j^x$. Looking back at Eqs. (52) and (54) we realize that this effective mean-filed Hamiltonian is equivalent to removing the α on Eq. (52) by plugging in the constrained minimum condition (54) and considering a uniform coupling $\lambda_j = \lambda$. Thus, in cases where $\mathcal{H}_S = \mathcal{H}_S^{\mathrm{MF}}$ the mean field effective Hamiltonian (84) is exact. This is because we are only applying mean field to the term arising from tracing out the light degrees of freedom. The mean field approximation decouples degrees of freedom by neglecting the fluctuations around the average value of the order parameter. Since we have proven that in the thermodynamic limit light and matter disentangle, and state vectors become separable, a mean field treatment of the light matter coupling becomes exact. Consequently, the effective spin-spin coupling that arises from it can also be treated exactly with mean field.

The *benefit of the mean field treatment* is twofold: as we will see shortly, it allows us to calculate how the coupling to the cavity modifies the bare spin properties; it also allows us to calculate observables, like m_x, in a simple manner. This is something that we could not do with our extension of Andolina and colleagues' method, which allowed us only to compute the phase boundary (if any), it also did not make explicit the modification of the spin properties. The mean field treatment is thus a complementary technique to gain qualitative insight of the model.

Before solving the Ising model it is interesting to establish a rigorous connection between the two order parameters that govern the model, namely m_x and α. Consider

$$\langle a \rangle = \alpha = Z^{-1} Tr_S \left(\int d^2\alpha \; \alpha \; e^{-\beta \mathcal{H}(\alpha)} \right). \tag{85}$$

Expanding this into real and imaginary integrals yields Gaussian integrals that can be computed, yielding

$$\alpha = Z^{-1} Tr_S \left(\frac{1}{\beta \omega_c} \frac{\lambda}{\omega_c \sqrt{N}} \sum_j \sigma_j^x e^{-\beta \mathcal{H}_{\mathrm{eff}}} \right) = \frac{\lambda}{\omega_c \sqrt{N}} m_x. \tag{86}$$

This confirms what we found in our extension of Andolina and colleagues' formalism [Cf. (54)]. It is also an important result because it proves that our two order parameters are not independent. A finite value of α implies a finite value of m_x. This may seem like a trivial assertion at this point, but it gains importance when we discuss models, such as the Ising model, in which a spontaneous symmetry breaking can occur independently of the coupling to the cavity. To better illustrate this point, let us solve now the ferromagnetic Ising model in transverse field.

$$\mathcal{H}_m = \frac{\omega_z}{2} \sum_j \sigma_j^x - \frac{J}{2} \sum_i \sigma_i^x \sigma_{i+1}^x. \tag{87}$$

A mean field approximation of $\mathcal{H}_S$ allows us to write

$$\mathcal{H}_{\mathrm{eff}}^{\mathrm{MF}} = \frac{\omega_z}{2} \sum_j \sigma_j^z - J m_x \sum_j \sigma_j^x + \frac{JN}{2} m_x^2 - \frac{2\lambda^2}{\omega_c} m_x \sum_j \sigma_j^x + \frac{\lambda^2 N}{\omega_c} m_x^2. \tag{88}$$

There are two alternative takeaways from this Hamiltonian, we can define $\lambda'^2 = \lambda^2 + \frac{J\omega_c}{2}$, in which case the resulting Hamiltonian is that of a plain Dicke model with an effective light-matter coupling constant λ':

$$\mathcal{H}_{\mathrm{eff}}^{\mathrm{MF}} = \frac{\omega_z}{2} \sum_j \sigma_j^z - \frac{2\lambda'^2}{\omega_c} m_x \sum_j \sigma_j^x + \frac{\lambda'^2 N}{\omega_c} m_x^2. \tag{89}$$

Alternatively we can define $J' = J + \frac{2\lambda^2}{\omega_c}$, in which case the resulting Hamiltonian will be that of a plain Ising model with an effective spin coupling J':

$$\mathcal{H}_{\mathrm{eff}}^{\mathrm{MF}} = \frac{\omega_z}{2} \sum_j \sigma_j^z - J' m_x \sum_j \sigma_j^x + \frac{J' N}{2} m_x^2. \tag{90}$$

This is remarkable, notice that the coupling to a single cavity is able to alter the properties of a macroscopically large number of electrons. The change in J is $\frac{2\lambda^2}{\omega_c}$, which depends on the collective coupling λ and not on the per-spin coupling $\lambda/\sqrt{N}$, this means that a relatively weak per spin coupling is sufficient to alter the Ising coupling constant. In experimental realizations in which many molecules are deposited in a single cavity, the per-spin coupling is usually poorly optimized, since the precise location of the molecules is not controlled. Hence the importance of depending exclusively on the collective coupling λ. The fact that the effective Hamiltonian can be expressed in terms of a mean field Ising model in transverse field also explains why the superradiant phase transition belongs to the universality class of the mean field Ising model, as we found in the early stages of this work (See Sec. 1.2).

It is convenient now to compute the free energy from Hamiltonian (89), and obtain the transverse magnetization variationally (See App. K for details).

$$\frac{F}{N} = \frac{\lambda'^2 m_x^2}{\omega_c} - k_B T \ln 2\cosh(\beta E) \qquad \text{with} \qquad E = \sqrt{\frac{\omega_z^2}{4} + \frac{4\lambda'^4 m_x^2}{\omega_c^2}}. \tag{91}$$

Minimizing with respect to the variational parameter m_x yields two possible solutions, either $m_x = 0$ corresponding to the disordered phase, or $\omega_c E = 2\lambda'^2 \tanh \beta E$, corresponding to the ordered phase. The latter is a transcendental equation that must be solved numerically to obtain the equilibrium value of m_x. The value of $\langle \sigma^x \rangle$ presented in Fig. 4 has been calculated this way, for the case of $J = 0 \implies \lambda' = \lambda$, i.e. for the plain Dicke model. In this case, it is clear that the symmetry breaking can originate exclusively from light-matter coupling, so a finite value of the order parameter m_x is indicative of the superradiant phase.

If we consider the addition of Ising coupling $J \neq 0$ a more subtle analysis is required, since a bare Ising model in transverse field can suffer a ferromagnetic phase transition by changing the coupling J. Thus, for values of $J > J_c$ past the critical coupling of the bare Ising model, which is $J_c^{\mathrm{MF}} = \omega_z/2$ in a mean field treatment, a finite value of m_x is no longer indicative of the superradiant phase transition, and instead it marks a ferromagnetic phase transition. This is corroborated by looking back at the expression of the effective light-matter coupling $\lambda^2 = \lambda'^2 - \frac{J\omega_c}{2}$, the critical condition of the effective Dicke model at $T = 0$ in this case is

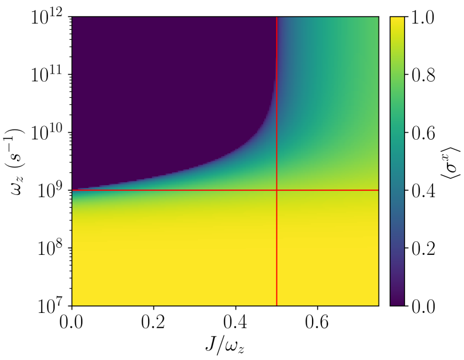

Fig. 5: Phase diagram for the modified mean field Ising model with a coupled cavity at $T = 0$ and $\rho = 5 \cdot 10^{20}\ cm^{-3}$. The colormap shows the transverse magnetization per spin $\langle \sigma^x \rangle$ at equilibrium. A finite value is indicative of the ordered phase. The vertical red line marks the critical value of the bare Ising constant J_c^{MF}, while the horizontal line marks the critical value of ω_z at which the bare Dicke model becomes superradiant for $\rho = 5 \cdot 10^{20}\ cm^{-3}$.

$$\lambda_c^2 = \frac{1}{4}\omega_z \omega_c - \frac{J\omega_c}{2}. \tag{92}$$

For the mean field critical value of $J_c^{\mathrm{MF}} = \omega_z/2$, the critical light-matter coupling λ_c goes to zero, indicating that light-matter coupling is no longer the driving factor inducing the phase transition. In Fig. 5 we show how the coupling to a single cavity affects the phase diagram of the mean field Ising model at $T = 0$. In the absence of cavity, we expect the model to undergo

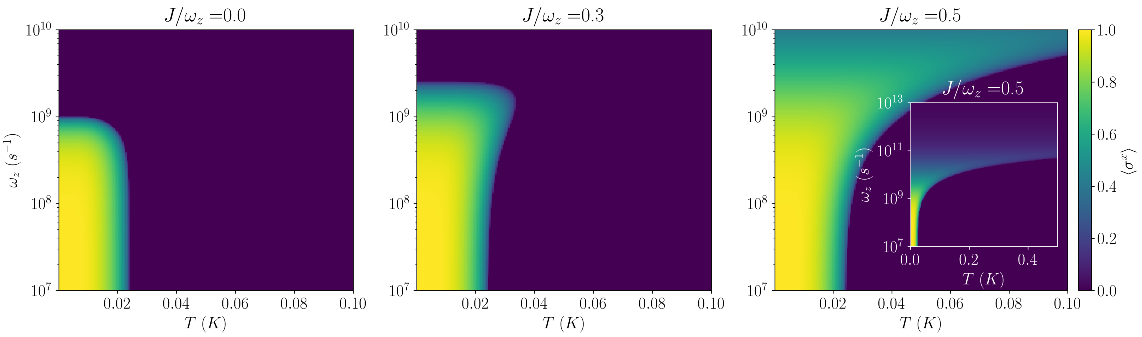

Fig. 6: Phase diagram for the modified Dicke model with Ising coupling in terms of ρ, T and ω_z. The colormap shows the transverse magnetization per spin $\langle\sigma^x\rangle$ at equilibrium. A finite value is indicative of the superradiant phase, provided $J/\omega_z < 1/2$. At $J/\omega_z = 1/2$ the Ising phase transition occurs and the phase boundary changes completely, indicating that the phase transition is no longer superradiant but ferromagnetic. This is better illustrated in the inset.

a phase transition at J_c^{MF} which is marked by a vertical red line. However, when the cavity is introduced, the effective coupling required to reach the ferromagnetic phase is modified, and ferromagnetism occurs for values of $J < J_c^{\mathrm{MF}}$. At the extreme, when ω_z reaches the critical value at which superradiance occurs in the bare Dicke model (indicated by a horizontal red line in Fig. 5), ferromagnetism occurs spontaneously even at $J = 0$. This is because the effective cavity-mediated coupling between spins is sufficiently intense to induce symmetry breaking on its own, without requiring direct spin-spin interaction. A complementary viewpoint is offered In Fig. 6, where we show how the Ising coupling facilitates the superradiant phase transition for values of $J < J_c^{\mathrm{MF}}$, this translates to a deformed phase boundary that makes the transition more resilient to both ω_z and T. In the same figure, we show as well that when the critical value of J_c^{MF} is reached the phase boundary changes its nature, indicating that the transition is no longer driven by light-matter coupling, but simply ferromagnetic. There is no longer a critical value of ω_z at which order breaks down.

These results can be compared with Fig. 7, where we show analogous results obtained with our extension of Andolina and colleague's method. Computing the critical condition in Eq. (70) requires knowledge of the spectrum of $\mathcal{H}_S$. We decided to tackle this issue numerically. In principle, we assumed that only low lying energy levels would have a significant contribution to the sum, allowing us to use optimized diagonalization routines that focus on the low energy sector of the spectrum. Unfortunately this intuition was flawed, because we were unable to obtain satisfactory results using Lanczos. We resorted then to naive exact diagonalization, which comes at the disadvantage of being able to solve much smaller systems, due to the exponential growth of the many-body Hilbert space. We found that a chain of 8 spins with periodic boundary conditions (PBC) provided a good trade off between numerical tractability and avoidance of finite size effects. In Fig. 7 we show the phase boundary obtained numerically, for a modified Dicke model with Ising coupling. The leftmost plot corresponds to $J = 0$ and is shown as a reference to contrast against Figs. 4 and 6. We find that the phase boundary coincides with that obtained with the original analytical solution of the Dicke model. To compare the center and rightmost plots of Figs. 6 and 7 an important distinction must be made: a mean field treatment of the Ising model provides a critical value of the Ising constant $J_c^{\mathrm{MF}} = \omega_z/2$ that deviates from the true critical value obtained with an exact analytical solution $J_c = \omega_z$ [26, Chap. 10]. This

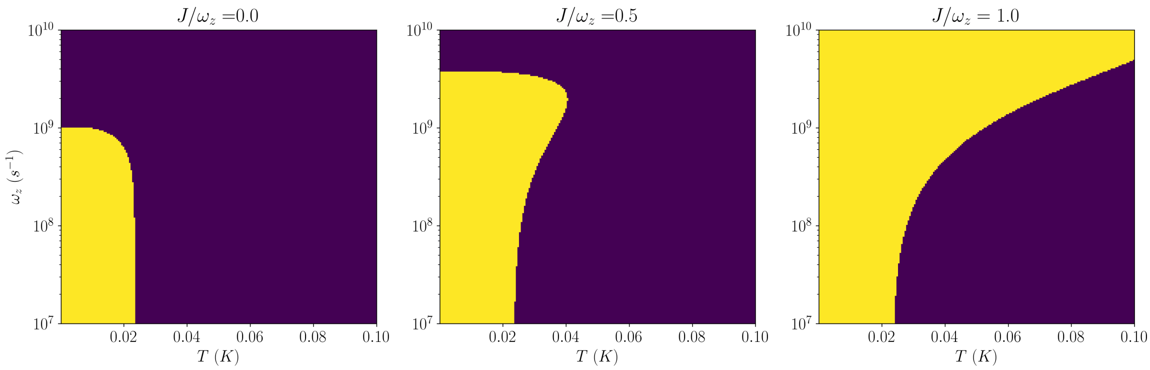

Fig. 7: Phase boundary of the modified Dicke model with Ising coupling for $\rho = 5 \cdot 10^{20} cm^{-3}$, computed with our extension of Andolina and colleagues' method for a chain of 8 sites with periodic boundary conditions. The yellow regions correspond to a finite value of α and thus of $\langle\sigma^x\rangle$, the purple regions corresponds to a zero value of the aforementioned observables.

explains why in Fig. 7 a value of $J = \omega_z$ is required to reach the point where the phase transition is simply ferromagnetic, in contrast with Fig. 6, where a value of $J = 0.5\omega_z$ suffices.

Some remarks are now in order. In the past few paragraphs we have discussed how the effective spin-spin coupling modifies the bare properties of the mean field Ising model, or, from an alternative viewpoint, the properties of the bare Dicke model. A lot of emphasis has been placed on distinguishing the phase transition occasioned by light-matter coupling, which occurs at $J < J_c$, from the phase transition occasioned by direct spin-spin interactions present in the bare Ising model, which occurs at $J \geq J_c$. This emphasis was motivated by our desire to provide a clear understanding of the driving factors behind the phase transition. However, from a practical point of view, the nature of the phase transition is irrelevant. In so far as Eqs. (68) and (86) show a relation between the expectation values of a and σ^x, the two "kinds" of phase transitions described lead to a finite population of photons in the cavity, *indicating photon condensation*.

3.4 Further generalizations of the Dicke model

There exist two more generalizations of the Dicke model that can be studied independently. They serve to generalize our findings to other setups that, we demonstrate, also contain the phase transition.

3.4.1 Spin $S > 1/2$

In order to reach the Dicke model, we had to assume that the electrons had no orbital angular momentum so the spin corresponded to the total angular momentum: $S = 1/2$. If we consider, e.g. molecular nanomagnets, they are described by a spin Hamiltonian with total spin S coupled to the magnetic field through the Zeeman term [24]. For simplicity, we consider here a isotropic spin-S

$$\mathcal{H} = \omega_z \sum_j S_j^z + \omega_c a^\dagger a + \frac{2}{\sqrt{N}}(a + a^\dagger) \sum_j \lambda_j S_j^x. \tag{93}$$

Where S_j^μ is the total spin operator along the μ axis. For a spin $1/2$ we have $S^\mu = \frac{1}{2}\sigma^\mu$ recovering our previous results. Repeating the treatment used before for the plain Dicke model [Cf. Eqs.

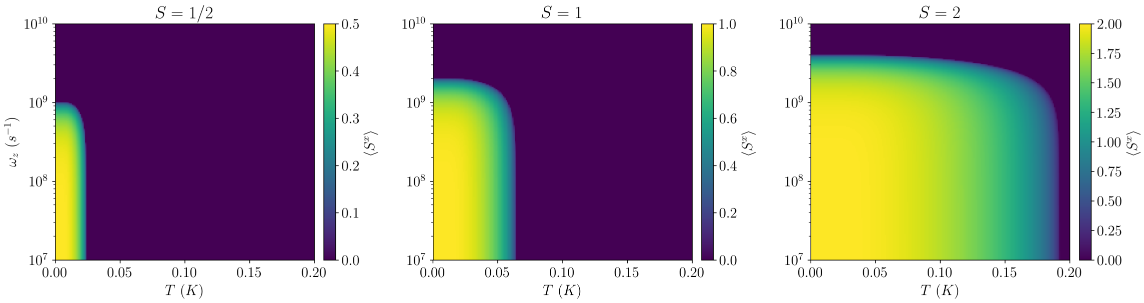

Fig. 8: Phase diagram for the modified Dicke model with spin S, for $\rho = 5 \cdot 10^{20} cm^{-3}$. The colormap shows the transverse magnetization per spin $\langle S^x \rangle$ at equilibrium. A finite value is indicative of the superradiant phase, where $\langle S^x \rangle \propto \alpha \neq 0$

(80) - (84)], we arrive at the effective Hamiltonian in mean field for spin S molecules

$$\mathcal{H}_{\text{eff}}^{\text{MF}} = \omega_z \sum_j S_j^z - \frac{8\lambda^2}{\omega_c} m_x \sum_j S_j^x + \frac{4\lambda^2 N}{\omega_c} m_x^2. \tag{94}$$

Where m_x is now defined as $m_x = \frac{1}{N}\sum_j S_j^x$. Like before, the value of m_x and the critical condition can be computed variationally. The free energy is found to be (See App. L for details on the calculation)

$$\frac{F}{N} = \frac{4\lambda^2 m_x^2}{\omega_c} - k_b T \ln\left[\frac{\sinh\left(\beta\frac{E}{2}(2S+1)\right)}{\sinh\left(\beta\frac{E}{2}\right)}\right]. \tag{95}$$

Where

$$E = \sqrt{\omega_z^2 + \left(\frac{8\lambda^2 m_x}{\omega_c}\right)^2}. \tag{96}$$

minimization of F/N with respect to m_x yields two possible solutions: either $m_x = 0$ corresponding to the subradiant phase, or

$$\omega_c E = 8\lambda^2\left[\frac{2S+1}{2}\coth\left(\beta E\frac{2S+1}{2}\right) - \frac{1}{2}\coth\left(\beta\frac{E}{2}\right)\right], \tag{97}$$

which is the trascendental equation that yields a finite value of m_x, signaling the superradiant phase. The critical condition is

$$\lambda_c^2 = \frac{1}{4}\omega_z\omega_c\left[(2S+1)\coth\left(\beta\frac{\omega_z}{2}(2S+1)\right) - \coth\left(\beta\frac{\omega_z}{2}\right)\right]^2. \tag{98}$$

In Fig. 8 we show the phase diagram for this model in comparison to the plain dicke model with $S = 1/2$, as we can see, the increase in spin has moderate effects that translate into a larger region of the parameter space being superradiant.

3.4.2 Multimode cavity

Our first assumption when quantizing the electromagnetic field in the Pauli equation was to consider that the cavity contained a single EM mode. This condition can also be relaxed without much alteration to the results obtained thus far. The corresponding Hamiltonian is

$$\mathcal{H} = \mathcal{H}_S + \sum_l^M \omega_l a_l^\dagger a_l + \sum_l \frac{\lambda_l}{\sqrt{N}}\left(a_l + a_l^\dagger\right)\sum_j^N \sigma_j^x. \tag{99}$$

The bounds found for Z (8) still apply here in most cases (See App. B.4 for an adapted proof to the multimode case) so we can substitute the bosonic operators for c-numbers, yielding

$$\mathcal{H}(\{\alpha_l\}) = \sum_l^M \omega_l |\alpha_l|^2 + \mathcal{H}_S + 2\sum_l \frac{\lambda_l}{\sqrt{N}} \mathrm{Re}(\alpha_l) \sum_j^N \sigma_j^x. \tag{100}$$

In an analogous fashion to what he have done, at this point, several times already throughout this work, we can compute the effective spin Hamiltonian for this model by tracing out the light degrees of freedom. In this case, we have to integrate over all the modes

$$\begin{aligned}
&\frac{1}{\pi}\int d^2\alpha_1 \dots \frac{1}{\pi}\int d^2\alpha_M \; e^{-\beta\mathcal{H}(\{\alpha_l\})} \propto \\
&\propto \prod_l^M \frac{1}{\pi}\int d\,\mathrm{Re}(\alpha_l) \exp\left[-\beta\left(\mathcal{H}_S + \omega_l Re(\alpha)^2 + 2\frac{\lambda_l}{\sqrt{N}}\mathrm{Re}(\alpha_l)\sum_j^N \sigma_j^x)\right)\right] \propto \\
&\propto \prod_l^M \exp\left[-\beta\left(\mathcal{H}_S - \frac{1}{N}\frac{\lambda_l^2}{\omega_l}\left(\sum_j^N \sigma_j^x\right)^2\right)\right].
\end{aligned} \tag{101}$$

So

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_S - \sum_l^M \frac{\lambda_l^2}{\omega_l}\frac{1}{N}\left(\sum_j^N \sigma_j^x\right)^2 \equiv \mathcal{H}_S - \frac{\lambda_{\text{eff}}^2}{\omega_{\text{eff}} N}\left(\sum_j^N \sigma_j^x\right)^2. \tag{102}$$

With

$$\frac{\lambda_{\text{eff}}^2}{\omega_{\text{eff}}} = \sum_l^M \frac{\lambda_l^2}{\omega_l}. \tag{103}$$

So we see that the resulting effective model is equivalent to the one obtained previously (83) for the case of a single-mode cavity, the only difference being that the coupling constant of the effective spin-spin coupling is now modified. Consequently, all results obtained so far apply to this model as well.

4 Experimental considerations

We have thus far shown that photon condensation is theoretically possible in magnetic QED. We have tested it analytically against multiple generalizations, proving that it is resilient against all of them. Hence, what is left at this point is to discuss the experimental setup required to produce and measure the phase transition.

In Fig. 3 we presented a schematic depiction of a superconducting cavity, in particular a CPW resonator, with deposited spins coupled to its magnetic field. We can see that the cavity is created by cutting the transmission line, forming two capacitors playing the role of mirrors in a Fabry-Perot cavity. The fundamental mode frequency is then $\omega_c = \frac{1}{\sqrt{lc}}\frac{\pi}{L_c}$, with l (c) the inductance (capacitance) per unit length of the transmission line and L_c the length of the cavity. The spins that interact with the cavity field are put there in the form of magnetic molecules containing free radicals. Unlike artificial (superconducting) atoms, these can be deposited in bulk, achieving densities in the range $2 - 9 \cdot 10^{20}\ cm^{-3}$ [25]. In order to measure the transition, we propose a transmission experiment. In such an experiment, a signal is sent through the central strip, it enters the cavity on one end, it is affected by the state of the system formed by the cavity and spins, and exits at the other end of the cavity. The input and output signals can then be measured, compared, and we can obtain information about the system's state. The resulting experimental setup can be understood as a Dicke model coupled to a transmission line, in which the coupling is modeled by a periodic driving of the cavity

$$\mathcal{H} = \frac{\omega_z}{2}\sum_j \sigma_j^z + \omega_c a^\dagger a + \frac{\lambda}{\sqrt{N}}(a + a^\dagger)\sum_j \sigma_j^x + f(t)(a + a^\dagger). \tag{104}$$

Where $f(t) = \epsilon e^{-i\omega t}$. The quantity to be measured in the experiment is the transmission, which is proportional to the susceptibility of the system to the periodic driving that is the signal.

$$t = -i\kappa\chi_\alpha \tag{105}$$

So we are interested in computing χ_α. For that, it is convenient to express the Hamiltonian in terms of total spin operators

$$\mathcal{H} = \frac{\omega_z}{2}S_z + \omega_c a^\dagger a + \frac{\lambda}{\sqrt{N}}(a + a^\dagger)S_x + f(t)(a + a^\dagger), \tag{106}$$

so we can write the equations of motion of the system in terms of these new observables. Presumably, the experiment would be carried out in a regime where the temperature can be considered to be zero, which simplifies the calculation. The equations of motion are the Heisenberg equations of motion ($\dot{\hat{O}} = i[\hat{\mathcal{H}}, \hat{O}]$) plus a phenomenological damping originating from the coupling to the transmission line

$$\begin{cases} \dot{S}_z = \frac{2\lambda}{\sqrt{N}}(a + a^\dagger)S_y - \gamma(S_z + N), \\ \dot{S}_x = -\omega_z - \gamma S_x, \\ \dot{S}_y = \omega_z S_x - 2\frac{\lambda}{\sqrt{N}}(a^\dagger + a)S_z - \gamma S_y, \\ \dot{a} = -i\omega_c a - i\frac{\lambda}{\sqrt{N}}S_x - if(t) - \kappa a. \end{cases} \tag{107}$$

Where γ and κ are, respectively, the damping constants for the spins and the cavity. If we consider $f(t)$ as a perturbation, i.e. $\epsilon \ll \omega_c$, we can write the average values of the operators in terms of their unperturbed values and the corresponding susceptibility: $\langle S_\mu \rangle = \langle S_\mu \rangle_0 + \epsilon e^{-i\omega t}\chi_\mu$, $\langle a \rangle = \alpha_0 + \epsilon e^{-i\omega t}\chi_\alpha$. Imposing the equilibrium condition: $\dot{S}_\mu = \dot{a} = 0$, we arrive to the system of equations that must be solved in order to obtain χ_α

$$\begin{cases} -i\omega\chi_z = \frac{2\lambda}{\sqrt{N}}\left(\chi_y(\alpha_0 + \alpha_0^*) + \langle S_y \rangle_0 (\chi_\alpha + \chi_\alpha)\right) - \gamma\chi_z, \\ -i\omega\chi_x = -\omega_z\chi_y - \gamma\chi_x, \\ -i\omega\chi_y = \omega_z\chi_x - 2\frac{\lambda}{\sqrt{N}}(\chi_z(\alpha_0 + \alpha_0^*) + \langle S_z \rangle_0 (\chi_\alpha + \chi_\alpha^*)) - \gamma\chi_y, \\ -i\omega\chi_\alpha = -i\omega_c\chi_\alpha - i\frac{\lambda}{\sqrt{N}}\chi_x - i - \kappa\chi_\alpha. \end{cases} \tag{108}$$

As reasoned in Sec. 1.2, we can assume α_0 and χ_α to be real. With that in mind, we can solve the system for each of the phases. In the subradiant phase, we have $\langle S_x \rangle_0 = -N$ and $\langle S_x \rangle_0 = \langle S_y \rangle_0 = \alpha_0 = 0$, which after some algebra (See App. M.1), yields

$$t = -\frac{i\kappa\left((\omega + i\gamma)^2 - \omega_z^2\right)}{(\omega - \omega_c)\left((\omega + i\gamma)^2 - \omega_z^2\right) - 4\lambda^2\omega_z + i\kappa\left((\omega + i\gamma)^2 - \omega_z^2\right)}. \tag{109}$$

In contrast, for the superradiant phase we have

$$\langle S_z \rangle_0 = -N\frac{\omega_z\omega_c}{4\lambda^2}; \quad \langle S_x \rangle_0 = N\sqrt{1 - \frac{\omega_z\omega_c}{4\lambda^2}}; \quad \langle S_y \rangle_0 = 0; \quad \alpha_0 = \frac{\lambda\sqrt{N}}{\omega_c}\sqrt{1 - \frac{\omega_z\omega_c}{4\lambda^2}}. \tag{110}$$

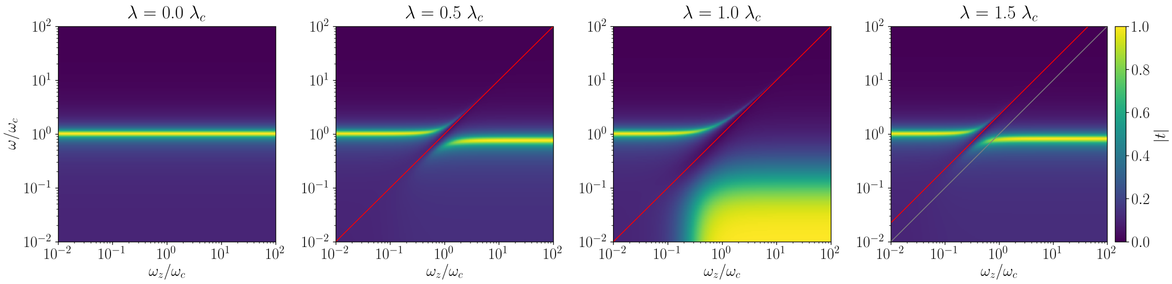

Fig. 9: Plots of the transmission as a function of the ratios between ω, ω_z and ω_c. The damping has been set to $\kappa = \gamma = 0.1$. From left to right we plot the diagram for: uncoupled cavity and spins, subradiant phase, right at the phase transition and finally in the superradiant phase. A red line marks the asymptote that breaks the resonance with the cavity. In the superradiant regime, the asymptote is displaced, this is indicated using a gray line that marks the position of the asymptote in the subradiant regime.

Solving for χ_α yields (See App. M.2)

$$t = -\frac{i\kappa\left((\omega + i\gamma)^2 - \frac{16\lambda^4}{\omega_c^2}\right)}{(\omega - \omega_c)\left((\omega + i\gamma)^2 - \frac{16\lambda^4}{\omega_c^2}\right) - \omega_z^2\omega_c + i\kappa\left((\omega + i\gamma)^2 - \frac{16\lambda^4}{\omega_c^2}\right)}. \tag{111}$$

In Fig. 9 we show the transmission as a function of ω, ω_c and ω_z in the different regimes of the system. For uncoupled qubits and cavity, the resonance occurs trivially at $\omega = \omega_c$. The inclusion of spin-cavity coupling breaks the resonance at $\omega_z = \omega_c$, since the numerator in Eq. (109) vanishes (ignoring damping) at this point. The transition is characterized by the disappearance of the $\omega_z > \omega_c$ resonant arm, which is instead replaced by a resonant region at $\omega \ll \omega_c$. This behavior can be explained by taking the appropriate limits in either Eq. (109) or Eq. (111). Recall that $\lambda_c = \frac{1}{4}\omega_c\omega_z$ and let us ignore the damping for simplicity. In the limit $\omega_z > \omega_c$, $\omega \ll \omega_c$, we have

$$t \approx \frac{i\kappa\omega_z^2}{\omega_c\omega_z^2 - \omega_c\omega_z^2 + i\kappa\omega_z^2} = 1. \tag{112}$$

In the superradiant phase the right resonant arm is restored but the asymptote is displaced. This can also be understood by looking at Eq. (111). Let us set $\lambda = \xi\lambda_c$ with $\xi > 1$, we see that the numerator vanishes (ignoring damping) at $\omega = \xi^4\omega_z$. This displacement of the asymptote is precisely what allows us to distinguish the sub- and superradiant phases in the transmission experiment.

Conclusions

In the first half of this Master's Thesis, we studied the problem of *equilibrium photon condensation* from a historical perspective. In Sec. 1, we have presented a thorough overview of the development of the topic until the present day. In the process, we learned a new technique to express the critical condition in terms of the susceptibility of the bare matter system [12], which we later generalized in the second block of our work. We also came across some recent uncontested publications supporting the case for a superradiant phase transition via *electric dipole coupling* and waveguide QED. We took the opportunity to respond to these contributions in Sec. 2, where come up with a rather straightforward no-go theorem in the dipole gauge, which we also connected to existing no-go theorems in the Coulomb gauge, providing a unified view of the impossibility of photon condensation with electric dipole coupling.

In the second half, we explored a new mechanism to achieve photon condensation, using magnetic coupling to escape previous no-go theorems. In Sec. 3 we successfully showed how to arrive to the Dicke model considering magnetic molecules. This allowed us to circumvent previous mechanisms impeding superradiance, such as the A^2 term. We explored the different generalizations of the Dicke model that can arise from our consideration of magnetic molecules, providing physical ranges for the parameters of the model at which photon condensation occurs. We did this with two complementary techniques: our generalization of Andolina and colleagues' method combined with exact diagonalization, and mean field theory. The latter gave us a clear interpretation of the influence of the cavity on the bare matter properties. Remarkably, it renormalizes the spin-spin coupling by an amount proportional to the collective coupling λ. Finally, in Sec. 4 we presented a transmission experiment designed to measure the transition. Our proposal is based on an architecture previously explored at QMAD where an ensemble of magnetic molecules is deposited in a CPW resonator coupled to a transmission line. We studied the transmission plots, showing that the sub- and superradiant phases can be distinguished with this setup.

This project is rather self contained, so possible theoretical continuations are scarce, besides publishing our findings. Nevertheless, it does have a clear continuation, which consists on presenting our results to the experimental team at QMAD and discussing with them the possibility of carrying out the experiment described in Sec. 4. We intend to do this as soon as possible.

References

[1] M. R. Matthews C. E. Wieman M. H. Anderson, J. R. Ensher and E. A. Cornell. Observation of bose-einstein condensation in a dilute atomic vapor. *Science*, 269(5221):198–201, 1995.

[2] E. R. Elliott C. Dutenhoffer J. R. Kellogg J. M. Kohel N. E. Lay K. Oudrhiri R. F. Shotwell Nan Yu D. C. Aveline, J. R. Williams and R. J. Thompson. Observation of bose–einstein condensates in an earth-orbiting research lab. *Nature*, 582(7811):193–197, 2020.

[3] J. Keeling and S. Kéna-Cohen. Bose–einstein condensation of exciton-polaritons in organic microcavities. *Annu. Rev. Phys. Chem*, 71(1):435–459, 2020.

[4] K. Hepp and E. H. Lieb. On the superradiant phase transition for molecules in a quantized radiation field: the dicke maser model. *Ann. Phys*, 76(2):360–404, 1973.

[5] J. Keeling P. Kirton, M. M. Roses and E. G. Dalla Torre. Introduction to the dicke model: From equilibrium to nonequilibrium, and vice versa. *Adv. Quantum. Techn*, 2(1-2):1800043, 2018.

[6] K. Wódkiewicz K. Rzażewski and W. Żakowicz. Phase transitions, two-level atoms, and theA2term. *Phys. Rev. Lett*, 35(7):432–434, 1975.

[7] P. Nataf and C. Ciuti. No-go theorem for superradiant quantum phase transitions in cavity QED and counter-example in circuit QED. *Nat. Commun*, 1(1), 2010.

[8] J. von Delft O. Viehmann and F. Marquardt. Superradiant phase transitions and the standard description of circuit qed. *Phys. Rev. Lett*, 107:113602, 2011.

[9] D. Hagenmüller and C. Ciuti. Cavity qed of the graphene cyclotron transition. *Phys. Rev. Lett*, 109:267403, 2012.

[10] V. Giovannetti L. Chirolli, M. Polini and A. H. MacDonald. Drude weight, cyclotron resonance, and the dicke model of graphene cavity qed. *Phys. Rev. Lett*, 109:267404, 2012.

[11] T. Grießer A. Vukics and P. Domokos. Elimination of the a-square problem from cavity qed. *Phys. Rev. Lett*, 112:073601, 2014.

[12] V. Giovannetti A. H. MacDonald G. M. Andolina, F. M. D. Pellegrino and M. Polini. Cavity quantum electrodynamics of strongly correlated electron systems: A no-go theorem for photon condensation. *Phys. Rev. B*, 100(12), 2019.

[13] A. Stokes and A. Nazir. Uniqueness of the phase transition in many-dipole cavity qed systems, 2019.

[14] R. M. White. *Quantum Theory of Magnetism.* Springer, 2006.

[15] S. Haroche and J. Raimond. *Exploring the Quantum.* Oxford University Press, 2006.

[16] R. H. Dicke. Coherence in spontaneous radiation processes. *Phys. Rev*, 93(1):99–110, 1954.

[17] Y. K. Wang and F. T. Hioe. Phase transition in the dicke model of superradiance. *Phys. Rev. A*, 7(3):831–836, 1973.

[18] K. Hepp and E. H. Lieb. Equilibrium statistical mechanics of matter interacting with the quantized radiation field. *Phys. Rev. A*, 8:2517–2525, 1973.

[19] E. H. Lieb. The classical limit of quantum spin systems. *Commun. Math. Phys*, 31(4):327–340, 1973.

[20] B. Simon. The classical limit of quantum partition functions. *Commun. Math. Phys*, 71(3):247–276, 1980.

[21] K. Inomata M. Bamba and Y. Nakamura. Superradiant phase transition in a superconducting circuit in thermal equilibrium. *Phys. Rev. Lett*, 117:173601, 2016.

[22] V. Giovannetti A. MacDonald G. Andolina, F. Pellegrino and M. Polini. Theory of photon condensation in a spatially-varying electromagnetic field, 2020.

[23] P. Simon D. Guerci and C. Mora. Superradiant phase transition in electronic systems and emergent topological phases, 2020.

[24] M. J. Martínez-Pérez J. García-Ripoll D. Zueco M. Jenkins, T. Hümmer and F. Luis. Coupling single-molecule magnets to quantum circuits. *New. J. Phys*, 15(9):095007, 2013.

[25] M. C. Pallarés-P. Hermosilla M. J. Martínez-Pérez M. D. Jenkins A. Angerer C. Sánchez-Azqueta D. Zueco J. Majer A. Lostao I. Gimeno, W. Kersten and F. Luis. Enhanced molecular spin-photon coupling at superconducting nanoconstrictions. *ACS Nano*, 2020.

[26] S. Sachdev. *Quantum Phase Transitions*. Cambridge University Press, 2011.

[27] J. L. García-Palacios, J. B. Gong, and F. Luis. Equilibrium susceptibilities of superparamagnets: longitudinal and transverse, quantum and classical. *J. Phys. Condens. Matter*, 21(45):456006, 2009.