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PERSPECTIVE

Quantum materials engineering by structured cavity vacuum fluctuations

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Abstract

A paradigm shift in the research of optical cavities is taking place, focusing on the properties of materials inside cavities. The possibility to affect changes of material groundstates with or without actual photon population inside cavities is an avenue that promises a novel view of materials science and provides a new knob to control quantum phenomena in materials. Here, we present three theoretical scenarios where such groundstate quantum phase transitions are predicted by the coupling of the matter to mere vacuum fluctuations of the cavity, as a realizations of cavity materials engineering in the dark.

1. Introduction

It has long been known in physics that when light is confined to very small volumes interesting phenomena can occur [1]. Most famously spontaneous emission amplified by light in cavities gives rise to the collective photon modes known as lasers [2, 3]. Since this discovery a rich tradition of research into optical cavities has developed with several groundbreaking and fundamental discoveries. For the present discussion it is particularly interesting that light-matter interactions inside optical cavities can be drastically enhanced [4], such that when matter is placed inside optical cavities quasiparticles of dual light-matter character can form, so called polaritons. A vast amount of results on these polaritons has been produced [5] and their formation and characterization is still being intensely studied with many challenges. For instance a big milestone in this line of research has been the realisation of polaritonic Bose–Einstein condensates [6, 7].

A recently developed idea has tried to shift the focus from the polaritons as a light-driven phenomenon towards what their formation does to the material that hosts them. In a pioneering field called polaritonic chemistry [8] the light-matter states are used to enhance and control chemical reactions. Formation of polaritons have, for example, been used to enhance the reaction pathway in molecules by changing the potential energy landscape [9–14]. Crucially, it was shown in polaritonic chemistry, that in the strong coupling regime already the vacuum fluctuations of the electromagnetic field in a cavity can couple to transitions of the electronic structure and hence novel induced phenomena can occur in dark cavities, i.e. in the absence of actual photons.

Analogously, engineering the cavity quantum-electrodynamical coupling to confined photon modes can impart a change to properties of quantum materials already via strongly coupling to vacuum fluctuations. This *vacuum cavity materials engineering* stands in contrast to the usual widely studied case of collective effects and condensation of driven (excited) polaritonic states. Formally, based on work done since early 2010's as part of two major projects funded by the European Research Council (DYNamo[5] and

[5] ERC-2010-AdG -Proposal No. 267374, 2011-2016.

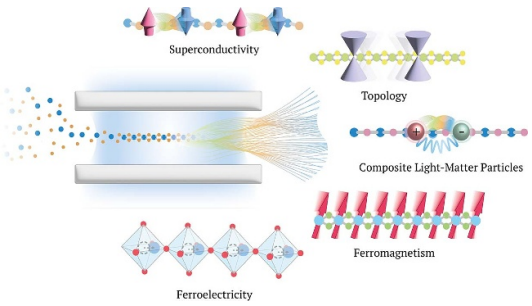

Figure 1. Materials properties engineering by cavity fluctuations. Schematics of light matter hybrid states are formed in an optical cavity affection electron, spin and phonon interaction. Examples of modifications of the ground state of a quantum materials are shown that can manifest in terms of change of the superconducting critical temperature, magnetic and electric order and the formation of polaritonic condensates.

QSpec-NewMat[6]) on the basic properties of strong light-matter coupled electronic structure [15–20], it was shown that quantum electrodynamics (QED) is bounded from below in the non-relativistic limit in contrast to relativistic QED, [21–25] i.e. even materials that are strongly coupled to light still have a true groundstate. Such a groundstate can be a mixed light and matter state. This development has led to a novel theoretical framework to describe strong light-matter phenomena in chemistry and materials science: quantum electrodynamical density functional theory [9, 17, 19, 26], which provides a unique and versatile *ab initio* approach to explore, predict, and control new states of matter in and out of equilibrium. This generalization of time-dependent DFT is the first method to explore the effects of addressing electronic states with photons while retaining the electronic properties of real materials.

The basic mechanism of groundstate modifications by the cavity is similar to many phenomena in condensed matter physics that are mediated by different bosonic fields (e.g phonons fluctuation in the standard BCS superconductors). Here, we are utilizing the vacuum fluctuations of the electromagnetic field in the cavity that is strongly coupled to virtual excitations in the material to modify properties of the material like superconductivity, magnetism, ferroelectricity, structural phase transitions, etc importantly, without the need of condensation [27]. Generally, photon-dressed groundstate phases do not necessarily have different properties than the material outside a cavity and often exciting changes to the material only manifest in its response properties [28, 29]. However, increasing the light-matter coupling in cavities can induce, if certain conditions are satisfied, a phase transition in a quantum material embedded in the cavity without external pumping, i.e. in a 'dark cavity'. It requires, however, that the cavity couples to a non-linearity in the material (e.g. a nonlinear phonon mode or a meta stable phase) and its description requires more than a single photonic cavity mode [4].

This approach is completely different from the research on polaritonic physics or driven quantum matter out of equilibrium [30], because the aim is not to dress collective excitations in the material with light, but to modify the groundstate and induce macroscopic changes in the material. Instead of supplying photons to reach condensation far from thermal equilibrium and/or a metastable state of matter, this approach strives to manipulate materials phenomena without photon excitation, but with dressing by the vacuum quantum fluctuations.

In the following we will discuss some recent works illustrating the evolution of this novel research area. We start with an example of cavity materials engineering of a ferroelectric phase in low temperature $SrTiO_3$ [31]. Then we discuss how cavities can be used to control correlated magnetic groundstates, as exemplified by α-$RuCl_3$, [32] and to modify the critical fluctuations at quantum phase transitions [33]. Furthermore, the quantum fluctuations of the vacuum can also lead to a modification the superconducting critical temperature [34], topological phase transition and the formation of novel quasiparticles [29] (see figure 1, not discussed below).

2. Cavity quantum materials

2.1. Photo-ferroelectricity

The perovskite $SrTiO_3$ (STO) is in its low temperature groundstate tetragonal phase a paraelectric, however, it has long been noted that at higher temperatures it displays a phonon mode which upon cooling softens and approaches zero energy at 4 K, where the material should become unstable and undergo a phase transition to

[6] ERC-2015-AdG -Proposal No. 694097, 2016-2021.

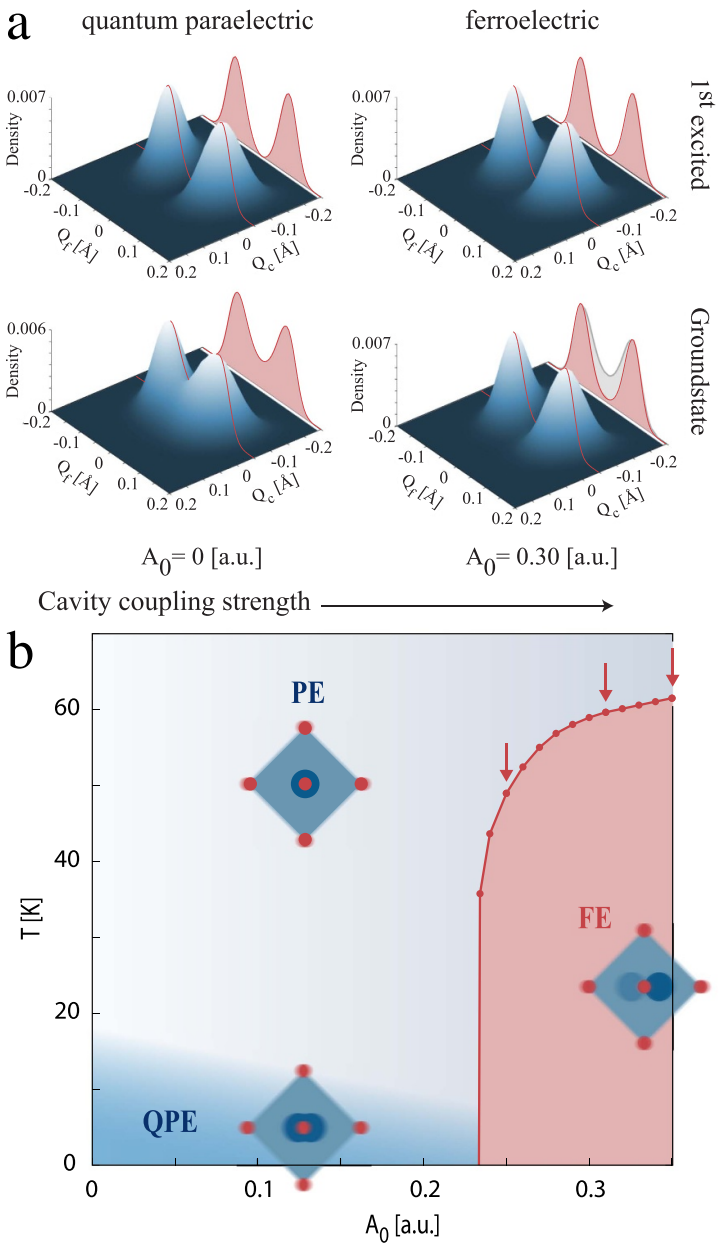

Figure 2. Ferroelectric-phase stabilized by by cavity vacuum fluctuations [31]. (a) Probability density of the ground- and first excited states of the low temperature tetragonal phase of STO for no ($A_0 = 0$) and finite cavity coupling. The delocalized groundstate prevents stable ferroelectricity for zero coupling, but for finite (strong) coupling the groundstate becomes degenerate with the first excited state and the lattice can localize in either of the two potential wells. (b) The cavity-temperature phase-diagram shows how the vacuum fluctuations of the cavity at zero temperature can stabilize the material against the quantum-paraelectric (QPE) phase by supporting the ferroelectric (FE) phase. At high temperatures both phases are overcome by the thermal excitations and the materials is a standard paraelectric (PE).

ferroelectricity like other materials of this family [35]. Instead of following this behaviour, the soft phonon mode in STO stabilizes to ∼0.5 THz and the material remains a paraelectric at low temperatures [36]. This behaviour has been explained by zero point fluctuations of the lattice, which prevents the formation of the long range macroscopic polarization associated with ferroelectricity. In other words, the quantum fluctuations of the lattice keep the material in an unpolarized phase, the *quantum* paraelectric phase [37].

By strongly coupling STO to a THz cavity mode, this low temperature behaviour can be changed. It has been shown from first principles calculations [31], that at low temperatures STO inside an optical cavity in the strong light-matter coupling regime stabilizes in a ferroelectric phase. Importantly, this stabilization is not a resonance effect but is due to the quantum fluctuations of the electromagnetic vacuum inside the cavity that strongly couples to the lattice. The vacuum fluctuations counteract the zero-point motion of the lattice. Figure 2(a) shows the first-principles density of the ground- and first excited state of STO in and outside a cavity. Such states reside in in a double well lattice potential. The quantum paraelectric state (at zero

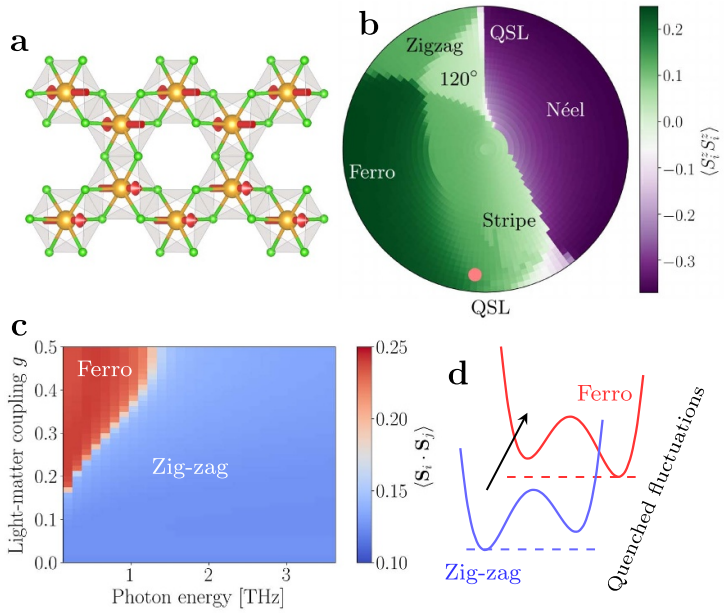

Figure 3. Magnetic phase transition in α-$RuCl_3$ induced by cavity vacuum fluctuations [32]. (a) Equilibrium magnetic structure of monolayer α-$RuCl_3$. The orange spheres denote Ru ions and the green spheres denote Cl ions. The zig–zag magnetic order, ferromagnetic within each chain and antiferromagnetic between chains, is indicated by the red arrows. (b) Magnetic phase diagram of the extended Kitaev model describing the low-energy order of α-$RuCl_3$. The three parameters are parameterized by $J = \cos\theta \sin\phi$, $K = \sin\theta \sin\phi$ and $\Gamma = \cos\phi$, with θ the polar angle and ϕ increasing from zero to π along the radial direction. The pink dot indicates the equilibrium state of α-$RuCl_3$. (c) Spin–spin correlation function $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ between nearest neighbor spins $\mathbf{S}_i$ and $\cdot\mathbf{S}_j$, indicating the transition from zig–zag to ferromagnetic order at low photon energies. (d) Phenomenological picture where the exact zig–zag groundstate, favored by magnetic fluctuations, is transformed into the ferromagnetic photo groundstate as magnetic fluctuations are quenched.

coupling to the cavity, $A_0 = 0$) can be seen to have a delocalized probability density in both potential wells, meaning that the states can fluctuate between the two wells, preventing the lattice from forming a stable ferroelectric polarization. Inside the cavity instead (for finite cavity-coupling strength A_0), the ground- and first-excited state density are equal and their energies are degenerate. The superposition of the degenerate bonding and anti-bonding states yields a state that is (spontaneously) localized in the right or left well, which is the microscopic condition for ferroelectric polarization. The localization is due to the vacuum fluctuations of electromagnetic field in the cavity, which are notably off-resonant from any lattice modes, in a process reminiscent of dynamical localization [38]. The groundstate of the material in the cavity is thus a nontrivial mixture of both electronic and photonic character. The coupling strength to the cavity then becomes a new dimension of the phase-space of the material, as shown in figure 1(b) for the cavity-temperature phase diagram. The vacuum fluctuations of the cavity compete with the thermal fluctuations of the lattice until for high temperature the material becomes a (normal) paraelectric.

2.2. Vacuum-driven magnetic phase transitions

The van der Waals antiferromagnet α-$RuCl_3$ consists of hexagonal layers of Ru ions, with each Ru surrounded by an octahedral cage of Cl ions (see figure 3). The symmetry breaking induced by the octahedral crystal field splits the previously degenerate Ru d-orbitals into two orbital sets, the t_{2g} and e_g manifold, separated by an energy Δ. The lower manifold is three-fold degenerate and contains the five valence electrons of the Ru^{3+} ion, as well as a single hole. Due to the competition of strong Coulomb interactions and spin–orbit coupling, the hole state forms an effective magnetic moment of magnitude $j = \frac{1}{2}$. The geometry of the material is such that the magnetic moments interact in a strongly anisotropic manner, with the spin Hamiltonian dominated by a ferromagnetic Kitaev interaction [39]. Due to the presence of multiple exchange paths, it is however supplemented by a weak Heisenberg interaction and additional anisotropic interactions [40]. In the ideal Kitaev limit, magnetic frustration is predicted to prevent long-range order down to zero temperature, resulting in a quantum spin liquid state with fractionalized excitations. More precisely, the elementary excitations are so-called spinons, which are fermionic and charge neutral quasi-particles [41]. The spinon dispersion vanishes linearly as $v|k|$ when $k \to 0$, but applying a weak out-of-plane magnetic field opens a gap at $k = 0$ and brings the system into a phase with topological order and non-Abelian excitations [42].

In α-$RuCl_3$, frustration and quantum fluctuations prevent magnetic ordering down to $T \approx 7$ K, below which the system orders into a zig–zag antiferromagnetic state (see figure 3(a)). The spin wave excitations are

gapless around the zig–zag ordering vector and display a gap of ≈2.5 meV at $k = 0$ [43]. Adjacent to the zig–zag state in the magnetic phase diagram is a ferromagnetic state (see figure 3(b)), differing in energy from the zig–zag state by only about 1 meV per unit cell. In fact, recent studies suggest that the classical groundstate of α-$RuCl_3$ is ferromagnetic, and that the zig–zag order is stabilized by magnetic quantum fluctuations [44]. This makes the phenomenology of α-$RuCl_3$ very similar to that of STO (cf section 2.1), such that a cavity-mediated quenching of the quantum fluctuations can be expected to bring about a change in the magnetic ground state.

A non-resonant coupling between the cavity and magnetic moments can be microscopically effected by coupling the cavity field to virtual electronic processes. In α-$RuCl_3$, the Kitaev interaction is found to mainly arise from hopping processes over the Cl ligands, while the sub-leading terms stem from direct Ru–Ru hopping. The relative strength of the magnetic interactions can therefore be controlled if the cavity dressing of these two exchange paths can be independently controlled. This is possible due to a different frequency scaling of direct and ligand mediated hopping processes, and a cavity operating in the few-THz regime is found to induce a transition from the zigzag to the ferromagnetic state [32]. In particular, the ratio of the direct exchange to the Kitaev interaction was found to be amplified by the cavity fluctuations. Since an enhancement of the exchange interaction suppresses magnetic fluctuations, the vacuum-induced phase transition can be readily interpreted within the picture presented above: a quench of magnetic fluctuations favors the ferromagnetic over the zig–zag antiferromagnetic state.

It should also be noted that the quantum spin liquid state can be stabilized over a large domain of cavity frequencies once the cavity is externally seeded into the few-photon regime [32]. Here, the magnetic transition is driven by drastic changes in the magnetic parameters, induced by near-resonant photon-assisted virtual electronic processes. Although such processes are suppressed in the vacuum limit, these results indicate the strong potential for a cavity to enhance light-matter coupling in a regime dominated by quantum fluctuations.

2.3. Cavity-renormalized quantum criticality

As demonstrated by both STO and α-$RuCl_3$, renormalizing material quantum fluctuations by the quantum vacuum fluctuations of an optical cavity can have a drastic effect on the equilibrium states of matter. However, inside a given phase the size of such fluctuations decay exponentially with the characteristic correlation length ξ, such that a significant light-matter coupling must be obtained on the scale of ξ to be relevant in finite systems. This situation becomes drastically different in the vicinity of a (quantum) critical point, where the correlation length diverges and fluctuations appear on every length scale. In particular, fluctuations of collective modes arising from a number of degrees of freedom on the order of $\xi \sim L$, with L the linear size of the system, can be expected to cancel the volume factor $g^2 \sim L^{-d}$ of the leading order light-matter coupling (where d is the spatial dimension).

The effect of a cavity on magnetic critical fluctuations was recently studied in a bilayer honeycomb antiferromagnet via large-scale quantum Monte Carlo simulations [33]. Many magnetic van der Waals materials crystallize in a honeycomb lattice and in particular the family of transition metal phosphorous trichalcogenides, MPX_3 with $M =$ Fe, Mn or Cr and $X =$ S or Se, all develop long-range order below some critical temperature T_c. For bilayers with an in-plane Neél antiferromagnetic order, as found in the compound $MnPSe_3$, a quantum phase transition can be induced by applying uniform pressure to tune the ratio of the inter-layer to intra-layer magnetic interactions to critical value $J_D/J \approx 1.64$ (see figure 4). This point marks the transition between a Neél ordered and dimerized antiferromagnetic state, and is described by an $O(3)$ critical theory.

For a critical system the main relevant properties are its critical exponents, and the position of the critical point. It was found in [33] that the cavity modifies the scaling behavior of the critical fluctuations by introducing finite-size scaling corrections (see figure 4). These corrections are of the form $A \sim A_0 + A_1 L^{1/\nu - d}$ for a singular observable, where A can be either the spin structure factor S or the magnetic susceptibility χ, and ν is the critical exponent of the correlation length. In the $O(3)$ universality class the exponent is negative, $1/\nu - d \approx -0.596 < 0$, such that the scaling correction vanishes in the thermodynamic limit. This behavior is well reproduced by a field theory obtained by coarse-graining the microscopic Hamiltonian, which shows that the scaling correction is a non-perturbative effect arising from a renormalization of the order parameter mass. Importantly, the field theory indicates that for different universality classes (such as the 1-D Potts class), the exponent can be positive and the correction diverge.

For the single-mode case investigated in [33], the critical point was found to be insensitive to the light-matter coupling. However, the effective field theory is straightforward to extend to the multi-mode case, allowing to assess the importance of beyond-dipole corrections, cf [4]. As expected from qualitative arguments, the main effect of including a finite-measure set of modes is to cancel the L^{-d} scaling of the light-matter coupling, leading to a finite effect in the thermodynamic limit. In particular, the field theoretic

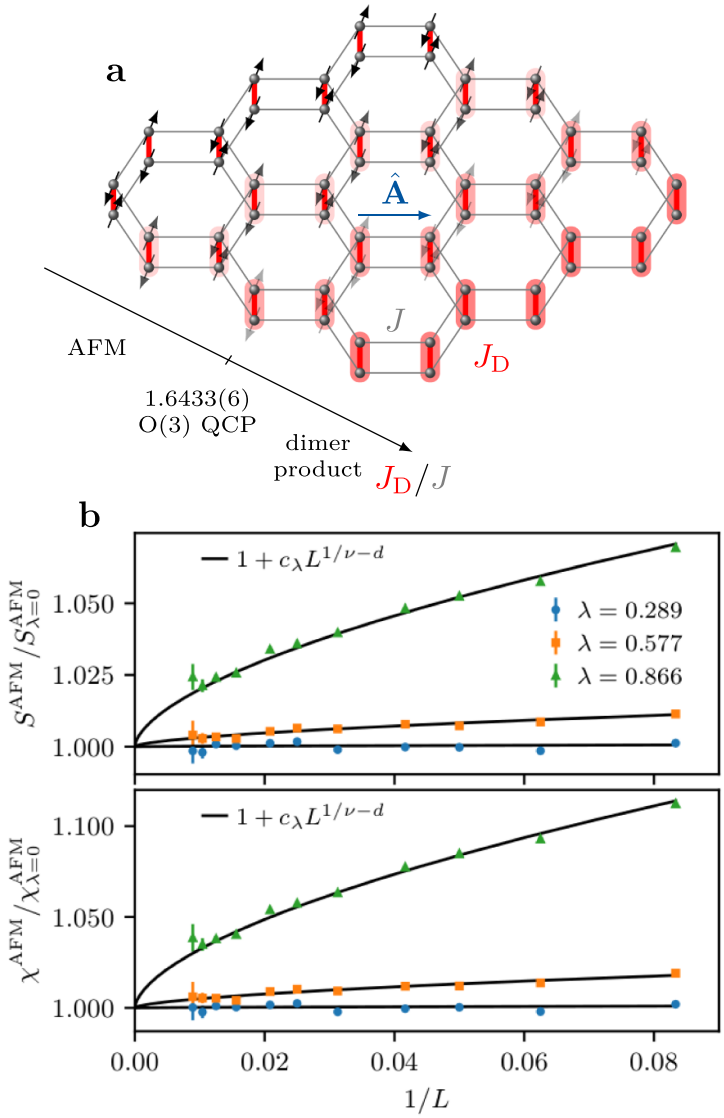

Figure 4. Scaling corrections of critical fluctuations in an antiferromagnetic honeycomb bilayer [33]. a, Magnetic ground state as a function of the ratio between inter-layer and intra-layer exchange couplings J and J_D. The system undergoes a quantum phase transition at the critical ratio $J_D/J \approx 1.64$. b, System size scaling of the critical fluctuations, here represented by the antiferromagnetic structure factor S_{AFM} and the antiferromagnetic susceptibility χ_{AFM}. The black lines are fits based on the effective field theory discussed in the main text.

approach predicts a finite shift of the quantum critical point in the multi-mode case, a prediction that is directly experimentally testable.

3. Outlook

QED-Cavity materials engineering is an exciting new field of research with the potential to transform the way we perceive and deploy materials science. Instead of searching for materials with specific properties, we might tune those properties across a range of values by tailoring quantum fluctuations of the material via their coupling to the cavity photon mode. Besides the mentioned examples, we envisage that cavity control of superconductivity, charge density waves, multiferroicity (among others, see figure 1) can be achieved. Importantly, there is still a lack of experiments demonstrating the potential of vacuum cavity engineering. First manifestations of such applications are the modification of the Quantum Hall transport [45] in a two-dimensional electron gas and the control of the metal-to-insulator transition temperature in TaS2[46]. The field is wide open for enterprising material scientists to explore the vast variety of possibilities and we expect many new phenomena to appear in the near future for quantum materials in cavities, catching up with the developments in chemistry where already clear effects on the groundstate have been shown experimentally [27, 47–49].

Data availability statement

No new data were created or analysed in this study.

Acknowledgments

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