Cavity Light-Matter Entanglement through Quantum Fluctuations

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The hybridization between light and matter forms the basis to achieve cavity control over quantum materials. In this work we investigate a cavity coupled to an XXZ quantum chain of interacting spinless fermions by numerically exact solutions and perturbative analytical expansions. We find two important effects: (i) Specific quantum fluctuations of the matter system play a pivotal role in achieving entanglement between light and matter; and (ii) in turn, light-matter entanglement is the key ingredient to modify electronic properties by the cavity. We hypothesize that quantum fluctuations of those matter operators to which the cavity modes couple are a general prerequisite for light-matter entanglement in the groundstate. Implications of our findings for light-matter-entangled phases, cavity-modified phase transitions in correlated systems, and measurement of light-matter entanglement through Kubo response functions are discussed.

Introduction.– Controlling material properties with light is a tantalizing avenue in condensed matter physics [1]. Notable recent achievements based on the ultrafast interaction of laser pulses with quantum materials include the light-induced anomalous Hall effect in graphene [2] and nonequilibrium superconducting-like states [3, 4]. Going beyond classical electromagnetic fields, materials properties can be influenced by hybridizing them strongly with the quantized electromagnetic field in optical resonators. An early example is the Purcell effect, where a change of the structure of the electromagnetic vacuum leads to a modified rate of spontaneous emission, or even Rabi oscillations [5]. More recently, the control of vacuum fluctuations and light-matter hybridization are utilized to design properties of extended solids, giving rise to a research field that has been coined "cavity quantum materials" [6–9]. Here coupling between quantum materials and cavity modes in a variety of cavity types and geometries can lead to the formation of hybrid polaritonic light-matter states with sometimes drastically modified many-body properties controlled by designing the electromagnetic environment [10–24].

The entanglement entropy between the light and matter parts of a system can be used as an indicator for the degree of their hybridization. Therefore the creation of light-matter entanglement is one of the key resources in the field. In the case of plain-vanilla polaritons, namely hybridized bosonic modes, [25] the generation of entanglement can be understood through models of coupled oscillators [26]. However in the growing field of (correlated) band electrons coupled to a cavity [19, 21, 22, 27–35] a clear picture of necessary and sufficient conditions for light-matter entanglement is still missing.

In this Letter we remedy this long-standing problem by showing that the quantum fluctuations of the current operator, which is generically the matter operator to which photonic gauge fields couple, play a pivotal role for achieving entanglement between correlated electrons and a cavity. By studying a paradigmatic and experimentally relevant model, we conclude that such fluctuations in fact constitute a necessary condition for non-zero light-matter entanglement. Additionally we find that a mean-field decoupling of light and matter, thus neglecting entanglement, yields qualitatively wrong results for photon related observables and does not reproduce the dominant corrections to fermionic correlation functions.

Model.– We consider a model of interacting spinless fermions coupled to the first transmittance resonance of a cavity fixing the wave vector perpendicular to the chain. In the direction of the chain, that we take as the x-direction, we only consider the spatially constant mode, i.e., wave vector $q_x = 0$. This amounts to the dipole approximation, which is justified for optical cavities since $c \gg v_{\mathrm{F}}$, where v_{F} is the Fermi velocity. We employ a quantized version of the Peierls substitution to couple light and matter in Coulomb gauge. This guarantees a gauge-invariant coupling for a low-energy theory [36, 37],

$$H = -t_h \sum_{j=1}^{L} \left(e^{i \frac{g}{\sqrt{L}} (a^\dagger + a)} c_j^\dagger c_{j+1} + h.c. \right) + U \sum_{j=1}^{L} \left(n_j - \frac{1}{2} \right) \left(n_{j+1} - \frac{1}{2} \right) + \Omega a^\dagger a. \tag{1}$$

Here, $c_j^{(\dagger)}$ annihilates (creates) a fermion at site j, and $n_j = c_j^\dagger c_j$ is the corresponding fermionic occupation. $a^{(\dagger)}$ annihilates (creates) a cavity photon, Ω denotes the bare cavity frequency, and g parametrizes the light-matter coupling. t_h denotes the nearest-neighbor hopping amplitude, U the nearest neighbor repulsion, and L the total number of sites.

We use periodic boundary conditions and a half-filled

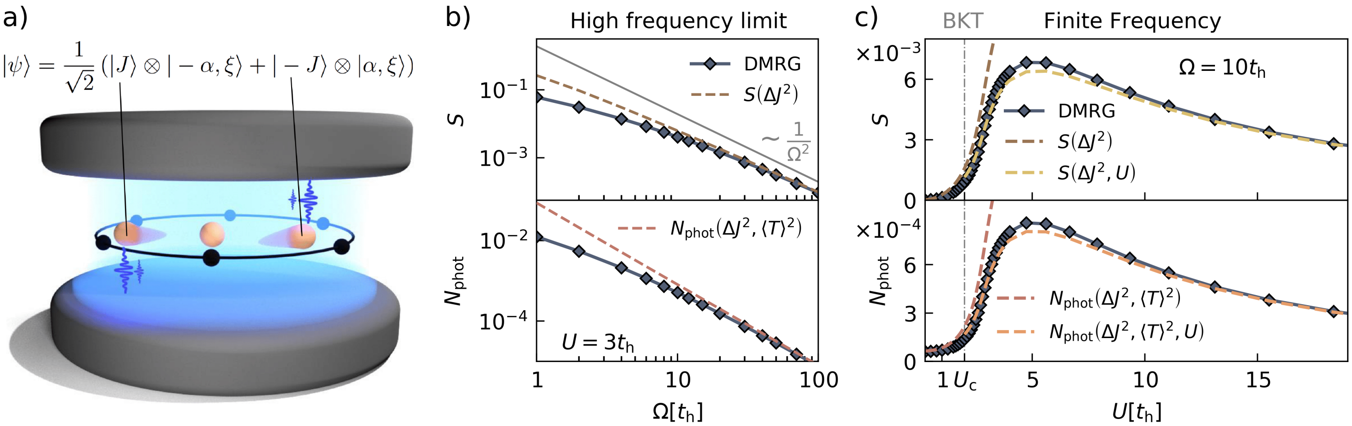

FIG. 1. **a)** Illustration of the mechanism generating light-matter entanglement in the ground state. While the expectation value of the current always vanishes $\langle J\rangle = 0$, the system is in a super-position of Bloch states with a well defined eigenvalue of the current operator J ($|\pm J\rangle$, only two are noted here for illustration) that each come with a certain squeezed coherent state $|\mp\alpha,\xi\rangle$ of the cavity mode Eq. (2) where the product $J\alpha$ has a fixed sign such that the overall inversion symmetry of the system is preserved. **b)** Entanglement entropy S (top) and photon number $N_{\rm phot}$ (bottom) as computed with DMRG (blue markers) as a function of the cavity frequency Ω; and as given by Eq. (7) $S(\Delta J^2)$ (top, brown dashed line) and Eq. (3) $N_{\rm phot}(\Delta J^2, \langle T\rangle^2)$ (bottom, red dashed line) in the high frequency limit. The gray line marks a $\frac{1}{\Omega^2}$ decay and we set $U = 3t_{\rm h}$ for the fermion-fermion interaction. **c)** Entanglement entropy S (top) and photon number $N_{\rm phot}$ (bottom) obtained with DMRG (blue marks) as function of U. Expressions obtained in the high-frequency limit Eq. (7) $S(\Delta J^2)$ (top, brown dashed line) and Eq. (3) $N_{\rm phot}(\Delta J^2, \langle T\rangle^2)$ (bottom, red dashed line) as well as those accounting for $U \sim \Omega$ Eq. (8) $S(\Delta J^2, U)$ (top, yellow dashed line) and Eq. (9) $N_{\rm phot}(\Delta J^2, \langle T\rangle^2, U)$ (bottom, orange dashed line) are overlayed. The frequency is set to $\Omega = 10t_{\rm h}$ in **c)**, the chain length to $L = 110$ and the light-matter interaction $g = 0.5$ throughout.

band. We set $e = \hbar = {\rm k_B} = {\rm c} = 1$ and the lattice constant $a_{\rm lat} = 1$. For $g = 0$ this model is equivalent to the XXZ spin Hamiltonian for which the exact groundstate can be obtained with the Bethe Ansatz [38]. At $U = 2t_{\rm h}$ the fermions undergo a quantum phase transition of the Berezinksii-Kosterlitz-Thouless (BKT) type from a Luttinger liquid (LL) phase to a charge-density wave (CDW) [39–43]. For $g > 0$ the fermionically noninteracting limit ($U = 0$) can be solved analytically and has a groundstate given by the product state of the unchanged Fermi sea for the fermions and a squeezed state for the cavity photon mode [31].

Analytical results.– First, we expand the Hamiltonian Eq. (1) to quadratic order in g. Employing a combined displacement and squeezing transformation of the Hamiltonian (see Supplemental Material [44]), we find a formal expression for the groundstate wave function of the system that is controlled in a high-frequency expansion,

$$|\Psi\rangle = \sum_u c_u |j_u, t_u\rangle \otimes |\alpha(j_u), \xi(t_u)\rangle, \tag{2}$$

where c_u are complex coefficients fulfilling $\sum_u |c_u|^2 = 1$, $|j_u, t_u\rangle$ are Bloch states composed of eigenstates of the current operator $J = \sum_k 2t_{\rm h}\sin(k)c_k^\dagger c_k$ and kinetic energy operator $T = -\sum_k 2t_{\rm h}\cos(k)c_k^\dagger c_k$, obeying $J|j_u\rangle = j_u|j_u\rangle$ and $T|t_u\rangle = t_u|t_u\rangle$, respectively. $|\alpha(j_u), \xi(t_u)\rangle$ are squeezed coherent states with displacement parameter $\alpha(j_u) = -\frac{g}{\sqrt{L\Omega}} j_u$ and squeezing parameter $\xi(t_u) = \frac{1}{2}\ln\left(1 - 2\frac{g^2}{\Omega}\frac{t_u}{L}\right)$. The expression Eq. (2) allows us to formally relate the photon number to expectation values of fermionic operators in the high-frequency limit

$$N_{\rm phot}(\Delta J^2, \langle T\rangle^2) = \langle\Psi|a^\dagger a|\Psi\rangle = \frac{g^2}{\Omega^2}\frac{\Delta J^2}{L} + \frac{g^4}{\Omega^2}\frac{\langle T\rangle^2}{L^2}. \tag{3}$$

To compute the entanglement entropy between light and matter we formally write the reduced density matrix of the photons $\rho_{\rm ph}$ by performing a partial trace of the fermionic degrees of freedom ${\rm Tr_f}$ on the full density matrix ρ, yielding

$$\rho_{\rm ph} = {\rm Tr_f}\,\rho = \sum_u |c_u|^2 |\alpha(j_u), \xi(t_u)\rangle\langle\alpha(j_u), \xi(t_u)|. \tag{4}$$

The corresponding entropy $S = -{\rm Tr_{ph}}\,\rho_{\rm ph}\ln(\rho_{\rm ph})$ quantifies the entanglement between light and matter. In the Supplemental Material [44] we show $S[\rho_{\rm ph}] \overset{L\to\infty}{\longrightarrow} S[\rho'_{\rm ph}]$ where

$$\rho'_{\rm ph} = \sum_u |c_u|^2 |\alpha(j_u)\rangle\langle\alpha(j_u)|, \tag{5}$$

i.e., the squeezing does not contribute to the entanglement in the thermodynamic limit. In the case of vanishing current fluctuations $\Delta J^2 = 0$ only a single coefficient c_u in Eq. (5) is different from zero leading to the reduced photonic density matrix of a pure state with vanishing entropy. We thus conclude the key results

$$\Delta J^2 = 0 \;\Rightarrow\; S = 0. \tag{6}$$

Hence current fluctuations are a necessary condition for light-matter entanglement. Computing the entanglement entropy between light and matter in the high frequency limit to leading order in $\frac{1}{\Omega}$ yields

$$S(\Delta J^2) = -\frac{1}{1+\chi}\ln\left(\frac{1}{1+\chi}\right) - \frac{\chi}{1+\chi}\ln\left(\frac{\chi}{1+\chi}\right)$$
$$\chi = \frac{g^2}{\Omega^2}\frac{\Delta J^2}{L}. \tag{7}$$

We note that $\chi = \frac{g^2}{\Omega^2}\frac{\Delta J^2}{L} \overset{L\to\infty}{\longrightarrow}$ const has a non-zero value in the thermodynamic limit.

Numerical solution.– We solve the model Eq. (1) including all orders of the gauge-invariant Peierls coupling [31, 37] with the density-matrix renormalization group (DMRG), using a photon number cutoff in the photonic part of the Hilbert space. For the implementation we use functions from the TeNPy library [45] and show further numerical details in the Supplemental Material [44].

The entanglement entropy S and photon number N_{phot} at $U = 3t_{\mathrm{h}}$ and $g = 0.5$ are shown as a function of the bare cavity frequency Ω in Fig. 1b. For comparison we also show the asymptotic high-frequency result obtained from Eqs. (7) and (3). We note that the population of the cavity stems solely from modified vacuum fluctuations and hence is small.

We now fix the frequency to an intermediate value of $\Omega = 10t_{\mathrm{h}}$. The light-matter coupling is set to $g = 0.5$. The entanglement entropy as a function of the fermion-fermion interaction U is shown in Fig. 1c. For $U \to 0$ the entanglement between light and matter vanishes together with the current fluctuations. We compare the result obtained with DMRG to Eq. (7) and find agreement for values of the interaction U up until the point where the interaction becomes comparable to the frequency at around $U = 3t_{\mathrm{h}}$. Beyond that point current fluctuations increase further eventually reaching a plateau ($\frac{\Delta J^2}{L} \to t_h^2, U \to \infty$) while the entanglement entropy decays as $\frac{1}{U^2}$ in the large interaction limit. We note that this does not contradict our previous statements around Eq. (6): current fluctuations are a necessary condition for light-matter entanglement but not a sufficient one.

We observe qualitatively similar behavior for the photon number N_{phot}, where the high-frequency result Eq. (3) compares well to the DMRG result for smaller interactions. One notable difference is that at $U = 0$ there is a finite photon number $N_{\mathrm{phot}} > 0$ due to a contribution from the squeezing of the photon states.

The behavior at larger interaction strength, $U > 3t_{\mathrm{h}}$, of the entanglement entropy and the photon number can be understood by noting that deep in the CDW phase, applying the light-matter coupling to the ground state wave function of the $g = 0$ system does not only create a photon excitation of energy Ω but also a charge excitation (through a hopping process) of energy U. To connect to the large U limit, we perform second-order perturbation theory in $\frac{t}{U}$ (see Supplemental Material [44] for details) and find that in order to account for this energy we effectively need to replace $\frac{1}{\Omega^2} \to \frac{1}{(\Omega+U)^2}$ in Eq. (3) and Eq. (7) such that we obtain

$$S(\Delta J^2, U) = \frac{-1}{1+\chi'}\ln\left(\frac{1}{1+\chi'}\right) - \frac{\chi'}{1+\chi'}\ln\left(\frac{\chi'}{1+\chi'}\right)$$
$$\chi' = \frac{g^2}{(\Omega+U)^2}\frac{\Delta J^2}{L}. \tag{8}$$

for the entanglement entropy and

$$N_{\mathrm{phot}}(\Delta J^2, \langle T\rangle^2, U) = \frac{g^2}{(\Omega+U)^2}\frac{\Delta J^2}{L} + \frac{g^4}{(\Omega+U)^2}\frac{\langle T\rangle^2}{L^2}. \tag{9}$$

for the photon number. We compare these expressions to the result obtained with DMRG for values of the interaction in the CDW phase $U > 2t_{\mathrm{h}}$ in Fig. 1c and find excellent agreement.

Comparison with mean-field approach.– Above we have found the necessity of current fluctuations in the fermionic system for light-matter entanglement in the coupled groundstate. In order to further elucidate the role of entanglement between light and matter for accurate results when $g \neq 0$ and $U \neq 0$, we compare the numerically exact DMRG results to a mean-field (MF) decoupling of light and matter that assumes a factorized groundstate wave function

$$|\Psi\rangle = |\psi\rangle_{\mathrm{f}} \otimes |\phi\rangle_{\mathrm{b}}. \tag{10}$$

Here $|\psi\rangle_{\mathrm{f}}$ is the fermionic part of the wave function while $|\phi\rangle_{\mathrm{b}}$ is the photon part. Importantly, both the cavity and matter systems are treated exactly by exact diagonalization and DMRG respectively, and only their interplay is approximated (for details see Supplemental Material [44]).

We compare the photon number as obtained with DMRG to that obtained with the MF approach in Fig. 2a. The DMRG result displays a peak in photon number after the BKT transition as a result of the interplay of current fluctuations and localization of the fermions at larger interaction in the CDW phase. The MF result agrees with DMRG at vanishing interaction $U = 0$ but from there on decreases monotonically as a function of increasing U, in stark contrast to the numerically exact DMRG result.

To obtain further insight into the specific wave functions obtained with the two methods, we plot the probability $P(n_{\mathrm{phot}})$ to find n_{phot} photons in the groundstate at $U = 3t_{\mathrm{h}}$. In MF one only finds even numbers of photons, $P(2n_{\mathrm{phot}}) > 0$ while $P(2n_{\mathrm{phot}} + 1) = 0$ for $n_{\mathrm{phot}} \in \mathbb{N}$. By contrast, the DMRG result shows a finite probability to find both even and odd photon numbers. We conclude that the MF decoupling captures

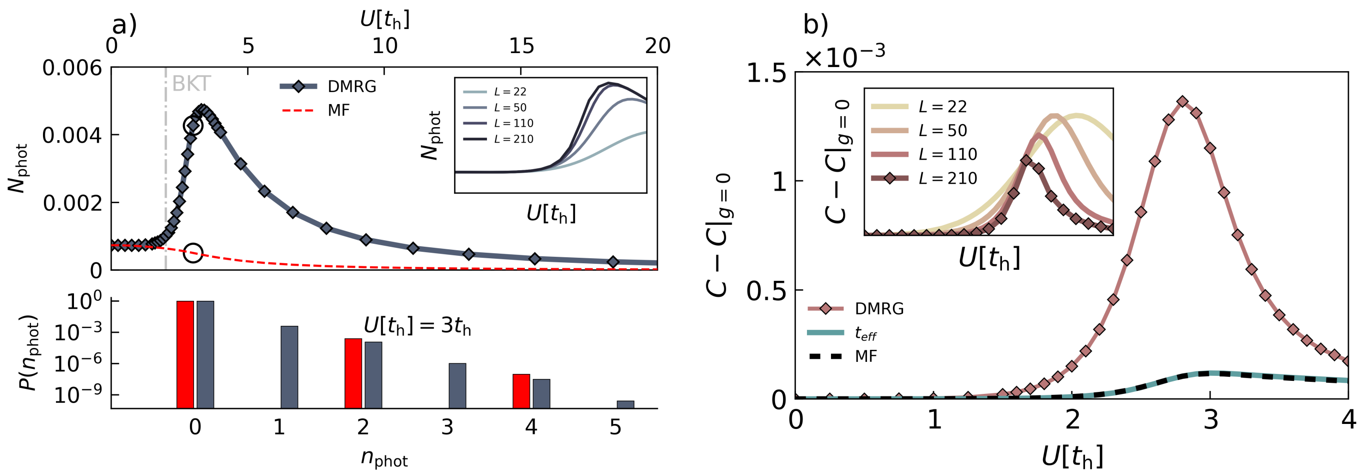

FIG. 2. **a)** Photon number N_{phot} as obtained with DMRG (dark blue points) and with a MF decoupling of light and matter (red dashed line) as a function of interaction strength U. The inset shows a finite size scaling of the photon number in the range $U = 0$ to $U = 4t_{\text{h}}$ as obtained in DMRG. The lower plot shows the probability to find n_{phot} photons in the ground state at $U = 3t_{\text{h}}$ (marked by circles in the upper part) as obtained with DMRG and the MF approach. **b)** Difference of long range correlations C (Eq. (11)) in the uncoupled system $g = 0$ and the coupled one $g = 0.3$ as obtained in DMRG (red points) and the mean field (MF) approach (black dashed line, also see Appendix **??**). We overlay the expected change of the correlator due to a change of the fermionic hopping t_{eff} (Eq. (12)) in blue that precisely matches the result from the MF approach but is at stark odds with the exact results. The inset shows a finite size scaling for the same quantity as obtained in DMRG. The chain-length is set to $L = 110$ and the frequency to $\Omega = 1t_{\text{h}}$.

the squeezing of the cavity mode (and thus $N_{\text{phot}}|_{U=0}$ matches the DMRG result) but fails to obtain the superposition of coherent states shown in Eq. (2). This is reasonable since the latter is mediated by the fluctuations of the current operator missed by the MF approach while fluctuations of the squeezing are suppressed in the TD limit also for the exact wave function such that the squeezing can be reproduced accurately.

We finally study the longest-ranged density-density correlations accessible in the L-site chain,

$$C := \langle n_0 n_{\frac{L}{2}} \rangle. \tag{11}$$

C serves as an order parameter for the BKT transition as $C = 0$ in the LL phase while $C > 0$ in the CDW phase, when $L \to \infty$. We show the result from both DMRG and the MF approach compared to that computed via DMRG in the uncoupled chain $g = 0$ in Fig. 2b. With DMRG we find cavity-increased correlations corresponding to an enhancement of the CDW over the LL phase. The MF approach produces a qualitatively similar result, however, with increases in correlations remaining approximately an order of magnitude smaller than those observed with DMRG. In fact we can fully understand the MF result from an effective rescaling of the hopping according to

$$t_{\text{eff}} = t\langle e^{ig\left(a^{\dagger}+a\right)}\rangle. \tag{12}$$

Since generally $t_{\text{eff}} < t$ the interaction U is effectively increased over the hopping leading to increased correlations. Yet, we show this heavily underestimates the way the cavity enhances effective correlations, and a truly light-matter entangled coupled wavefunction is required to obtain the full picture.

Finally we perform a finite-size scaling of the changes in C and find a monotonic decay as the system size L increases. The reason for this vanishing influence of the cavity on the correlator in the thermodynamic limit lies in the fact that we have approximated the cavity degrees of freedom with a single bosonic mode, which has vanishing energy density in the thermodynamic limit.

Discussion.– In this work we have shown that quantum fluctuations of the current operator are a necessary condition for light-matter entanglement between spinless fermions in an interacting quantum chain and a cavity mode. This result is expected to hold in more general settings including higher dimensions, different forms of the interaction, and the inclusion of a spin degree of freedom. The reason is that for the analytical calculations performed here, neither dimensionality nor the particular form of the interaction played any role whatsoever. We used the dipole approximation which is expected to hold in the far field in optical cavities since $c \gg v_{\text{F}}$. This assumption might, however, be violated in plasmonic cavities and in the presence of strong near-field effects. The fluctuations of the current operator can in principle be directly measured through Kubo response functions [46], possibly providing a quantum-metrological handle to measure light-matter entanglement [47, 48]. This is reminiscent of the recent finding that the quantum Fisher information, that is directly related to the quantum fluctuations of an operator [48], is a multipartite entanglement witness in [49] and out of thermal equilibrium [50]. However, we point out that the relations found in this work establish quantum fluctuations as a necessary condition for entanglement and not a sufficient one, as would be required [51] for quantum fluctuations to serve as a complete entanglement witness. We note that the role of quantum fluctuations for light-matter entanglement in cavity quantum materials at finite temperature, where also thermal fluctuations are present, and out of thermal equilibrium are interesting subjects that deserve further study.

Additionally we showed that the entanglement between light and matter can lead to increased effective correlations. Finally we demonstrated by finite-size scaling that a single cavity mode can only influence matter properties in mesoscopic, but not macroscopic systems. The inclusion of macroscopically many modes, that are naturally present in certain experimental setups, could for instance enable the creation of long-ranged charge correlations in a regime where correlations are otherwise only short-ranged. Such an effect might explain the recent experiment in which a Fabry-Perot resonator modified the metal-to-CDW-insulator transition in 1T-TaS_2 [27]. On the flip side, the measurement of modifications of both electronic and photonic properties in hybrid systems that cannot be understood from a product wave function alone might point us to finding entanglement in cavity-matter systems.

DATA AND CODE AVAILABILITY

Data included in the paper are available upon request, the codes used to generate it are openly available at `https://github.com/GiacomoPassetti/Paper_XXZ_cavity.git`.

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