Ultrastrong time-dependent light-matter interactions are gauge-relative

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Time-dependent Hamiltonians are a widespread means by which to describe controllable experimental operations. In particular, control over light-matter interactions is vital for numerous applications. Due to gauge-freedom, there are many different definitions of light and matter as quantum subsystems. These definitions only coincide when the interaction vanishes. Correspondingly, various forms of interaction are commonly found within the literature. In contrast, justification for using a particular form is seldom found. We show that in the absence of an argument to choose a particular gauge when promoting the coupling parameter to a time-dependent function, the description that results is essentially ambiguous. For sufficiently strong and non-adiabatic interactions, the qualitative physical predictions of final subsystem properties, such as entanglement and photon number, depend on the gauge chosen. This occurs even when the coupling vanishes at the preparation and measurement stages of the protocol, at which times the subsystems are unique and experimentally addressable. These findings are important for all situations in which specific properties are sought through fast interaction switching, as may occur for example, within quantum information, communication, metrology, simulation, and control.

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I. INTRODUCTION

Exploiting controlled light-matter coupling is important for quantum computation [1–4], quantum communication [5], quantum metrology [6], and quantum simulation [7, 8]. In the search for scalable platforms operating at room-temperature, strong light-matter coupling has become of major interest through solid-state systems, such as semiconductor quantum wells [9] and dots [10, 11], through two-dimensional [12] and organic [13] materials, and through superconducting circuits [14–18]. For most applications it is necessary to realise and/or manipulate specific quantum states via Hamiltonians with tunable parameters [17, 19–27]. For example, time-dependent light-matter coupling in cavity QED can be used to realise a universal set of gates for quantum information processing [28]. Time-dependent couplings are also routinely found in models of driven systems, such as the extended Rabi-model, which is important in optimal control theory [29–32].

In these applications properties such as entanglement, which pertains to the subsystems of the overall light-matter composite, are of central importance. A key concept in the study of such properties is locality. An observable is said to be local with respect to a bipartition $\mathcal{H}_A \otimes \mathcal{H}_B$ of a composite system $AB$, if it has the form $I_A \otimes B$ whereby it is associated with subsystem $B$ only, or if it has the form $A \otimes I_B$ whereby it is associated with subsystem $A$ only. Locally equivalent Hilbert space bipartitions $\mathcal{H}_A \otimes \mathcal{H}_B$ and $\mathcal{H}_A' \otimes \mathcal{H}_B'$ are connected by local unitary transformations, i.e., unitary transformations that have the form $U_A \otimes U_B$ [19, 33].

In QED tensor-product bipartitions of the state space are induced by canonical operators. This is highly non-trivial, because the canonical momenta are not manifestly gauge-invariant. For example the Maxwell field canonical momentum $\Pi$ in the Coulomb gauge coincides with the gauge-invariant transverse electric field $\Pi = -E_T$ but in the Poincaré (multipolar) gauge it coincides with the gauge-invariant transverse displacement field $\Pi = -D_T$ [34]. These fields differ by the transverse polarisation $P_T = D_T - E_T$, which only vanishes when the light-matter coupling vanishes. To see how this effects notions of “light” and “matter” as quantum subsystems it suffices to note that the bosonic annihilation operator $a_\lambda(k)$ for a “photon” with polarisation $\lambda$ and momentum $k$ is defined directly in terms of the Fourier transform of the canonical field $\Pi(x)$ [34]. This dependence on $\Pi$ immediately implies that for non-vanishing coupling the term “photon” refers to physically different bosonic quanta in the Coulomb and Poincaré gauges.

The topic of dividing light-matter systems into subsystems by selecting a particular interaction Hamiltonian has received considerable attention over the past seven decades. Focus has predominantly been placed on comparing the Coulomb and multipolar gauge interactions, with only a limited few exceptions [35–39]. A notable exception is the so-called Pauli-Fierz representation, which attempts to isolate the component of the electromagnetic field that is tied to the material system. This representation has been used in calculating radiative corrections such as the Lamb-shift [40]. As well as having considered only a handful of representations, previous studies have invariably focussed specifically on establishing gauge-invariance of the $S$-matrix [34, 41–47], or else have considered the natural lineshape problem of spontaneous emission in conventional weak-coupling and Markovian

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regimes [36, 37, 48–52]. It is now well-known that QED $S$-matrix elements calculated by treating the interaction as a weak perturbation are in fact gauge-invariant at every order of perturbation [34, 46, 47]. However, this result is physically limited. Indeed, the gauge-invariance of the $S$-matrix is a direct consequence of a property specific to scattering processes. Namely, that the interaction is switched on adiabatically from $t = -\infty$ and then switched off adiabatically, vanishing at $t = +\infty$ [34]. In non-truncated models and beyond scattering theory, there appears to be no general consensus as to which definitions of light and matter are most appropriate in which situations. Coulomb and multipolar gauge formulations seem to be used with equal frequency, but without justification as to why a particular form has been chosen. This is usually unproblematic within the conventional regime of sufficiently weak and slow interactions, because differences in subsystem predictions resulting from different theoretical definitions are typically negligibly small. On the other hand, significant attention has not so far been given to the availability of different subsystem definitions in the context of the faster and stronger time-dependent couplings that are required for applications.

More recently, a generalised formulation of QED has been developed, whereby the choice of gauge is controlled using a classical function [39, 46, 56]. As a special case of this the gauge can be controlled using only a single real parameter $\alpha$ [36, 38, 39]. With this formalism it has been shown that every gauge-fixing transformation is coupling-dependent and non-local with respect to every decomposition of the system into “matter” and “light” quantum subsystems. In the $\alpha$-gauge framework, each $\alpha$ is associated with its own set of canonical operators, which induces a corresponding state space bipartition $\mathcal{H}_{m}^\alpha \otimes \mathcal{H}_{l}^\alpha$. Bipartitions labelled by distinct values of $\alpha$ are not locally equivalent. This cements the conclusion that theoretical QED subsystems can only be defined relative to a choice of gauge, a feature we refer to as subsystem gauge-relativity. The $\alpha$-gauge framework includes the Coulomb and multipolar gauges as special cases, but also includes a third noteworthy representation in which number non-conserving interactions are eliminated from the linear part of the interaction Hamiltonian. This representation can be interpreted as one in which the cloud of virtual photons tied to the ground state of the bare material system, has been absorbed into its definition.

In general the most relevant decomposition of a system’s Hilbert space is induced by the experimentally accessible observables, including interactions and measurements [19, 33, 57–59]. For example, with respect to distinct Hilbert space bipartitions a two qubit Bell-state could be maximally entangled or not entangled at all [59, 60]. It is not physically meaningful to refer to this state as being entangled, unless one can also specify how its constituent physical degrees of freedom can be manipulated or probed. Thus, physical subsystems and their properties must be specified relative to a particular set of experimental capabilities.

There have been a number of recent works which consider the breakdown of gauge-invariance due to a two-level approximation of the matter system [39, 53–55]. Here we identify a different mechanism by which ambiguity can enter the theory due to gauge-freedom. We consider the treatment of tuneable light-matter interactions through the promotion of the coupling to a time-dependent function. Unlike the two-level approximation, this procedure does not ruin the unitarity of gauge-transformations. We show nonetheless, that for sufficiently strong light-matter interactions, final subsystem properties, such as entanglement and subsystem energies, depend significantly on the definitions of light and matter adopted during their interaction. This occurs even if the interaction is not present at the initial and final stages of the protocol, at which times the subsystems are uniquely defined and can be individually addressed.

Our results are cautionary in nature. We show that in strong-coupling non-adiabatic regimes, unless a justification for using a particular form of light-matter coupling can be provided within the context of the experiment being modelled, then even qualitative physical conclusions drawn from the theory are unreliable. We consider the predicted final energy and information exchange between light and matter, after their interaction has been switched on and then off again non-adiabatically. We find that different qualitative physical regimes of energy exchange can be realised by varying nothing but the gauge in which the interaction is modelled. Our findings are of major importance for the numerous applications whose basic aim is to generate specific properties, such as entanglement or a certain number of photons. We frame our findings within the context of cavity QED, but note that the results obtained are a general feature of QED, which in turn underpins the description of all light-matter systems. We remark also that sub-cycle, ultrastrong light-matter interaction switching has already been achieved within semiconductor micro-cavity QED systems [15]. These systems offer a promising experimental platform to probe the unique and fundamental aspects of ultrastrong non-adiabatic light-matter physics that we report.

II. LIGHT MATTER HAMILTONIAN IN AN ARBITRARY GAUGE

We present our approach within the context of cavity QED. Our aim is to determine the dependence of local light and matter properties on the chosen definitions of the light and matter subsystems during their interaction. It suffices for this purpose to consider a simple material system for which analytic expressions may be obtained. We therefore consider a bound electron $-e$ with position $r$ confined in all spatial dimensions except the direction $\xi$ of the polarisation of a single cavity-mode, in which it is bound harmonically. The mode is described by the components $A$ of the gauge-invariant transverse vector
potential along the direction $\varepsilon$. We adopt a generalised approach in which the choice of gauge is determined by a real parameter $\alpha$. The energy is derived from first principles and is found to be [39]

$$
H = E_{\text{matter}} + E_{\text{cavity}},
$$

where the matter and cavity energies are in turn the sum of kinetic and potential energies; $E_{\text{matter}} = m(r^2 + \omega_m^2 r^2)/2$ and $E_{\text{cavity}} = v(\dot{A}^2 + \omega^2 A^2)/2$. Here $v$ denotes the cavity volume and $\omega$ denotes the cavity frequency while $\omega_m$ denotes the frequency of the matter oscillator. The above expression for the energy is manifestly gauge-invariant ($\alpha$-independent). However, the material and cavity canonical momenta $p$ and $\Pi$ are manifestly gauge-dependent and are given by [39]

$$
p = m\dot{r} - e(1 - \alpha)A, \quad \Pi = \dot{A} + e\alpha \frac{r}{v}.
$$

Only when $e = 0$ are the canonical operators unique ($\alpha$-independent) in which case the Hamiltonian in Eq. (1) simply describes non-interacting light and matter. Notice that the above two equations constitute a superposition of distinct minimal-coupling prescriptions. The additional term in the minimal coupling prescription $p \rightarrow p + eA$ is weighted by $1 - \alpha$ and the additional term in the minimal-coupling prescription $\Pi \rightarrow \Pi - ev/v$ is weighted by $\alpha$.

The material and cavity canonical operators satisfy the canonical commutation relations $[r, p] = i$ and $[A, \Pi] = i/v$ while all other commutators between canonical operators vanish. Cavity and matter bosonic ladder operators can be defined in the usual way as $a = \sqrt{v/2\omega}(\omega A + i\Pi)$ and $b = \sqrt{1/2m\omega_m(m\omega_mr + ip)}$ respectively, with $[a, a^+] = 1 = [b, b^+]$. By expressing the energy in Eq. (1) in terms of canonical operators using Eqs. (2), and subsequently expressing the canonical operators in terms of ladder operators one obtains the Hamiltonian $H^\alpha = H_0 + V^\alpha$ where $H_0 = \omega(a^\dagger a + 1/2) + \omega_m(b^\dagger b + 1/2)$ and

$$
V^\alpha = \frac{\eta^2\omega}{4} \left[ (1 - \alpha)^2(a^\dagger + a)^2 + \delta\alpha^2(b^\dagger + b)^2 \right]
+ i\alpha u_\alpha^-(ab^\dagger - a^\dagger b) + i\alpha u_\alpha^+(a^\dagger b^\dagger - ab)
$$

with $\eta = e/\sqrt{\omega v}v$ a dimensionless coupling parameter, $\delta = \omega/\omega_m$, and $u_\alpha^\pm = \eta\omega_m\sqrt{[1 - \alpha \mp \delta\alpha]/2}$. The Coulomb and Poincaré (multipolar) gauges that are commonly encountered in atomic physics are special cases obtained by choosing $\alpha = 0$ and $\alpha = 1$ respectively. We note that although the quadratic terms on the top line of Eq. (3) could be removed via local Bogoliubov transformations, this would give rise to coupling-dependent renormalised matter and cavity frequencies in the free part of the Hamiltonian. In what follows we will be dealing with time-dependent couplings for which the interaction should be defined as the component of the Hamiltonian that depends on the coupling parameter, as in Eq. (3).

Hamiltonians associated with distinct gauges $\alpha$ and $\alpha'$ are related by $H^{\alpha'} = R_{\alpha\alpha'} H^\alpha R_{\alpha'\alpha}$ where

$$
R_{\alpha\alpha'} = \exp[-ie(\alpha - \alpha')r \otimes A]
$$

is a non-local unitary gauge-fixing transformation. Since according to Eq. (4) gauge transformations are unitary the theory is strictly gauge-invariant; predictions for any given observable can be found in any gauge and these predictions are unique. However, since gauge-transformations are non-local, for distinct gauges the quantum subsystems are defined in terms of different gauge-invariant observables, that is, the subsystems can only be defined relative to a choice of gauge. The theory itself does not favour the use of a particular set of canonical operators over any other, each set defines a pair of mathematically valid theoretical subsystems.

The bosonic operators $a$ and $b$ are implicitly $\alpha$-dependent due to their dependence on the canonical momenta, which represent different observables for different $\alpha$. In the gauge $\alpha$ the number eigenstates of the material operator $b^\dagger b$ span the material Hilbert space $H^\alpha_m$ while those of the cavity operator $a^\dagger a$ span the cavity space $H^\alpha_l$. When combined these spaces make up the composite state space $H^\alpha = H^\alpha_m \otimes H^\alpha_l$. Although this space is physically unique inasmuch as $H^{\alpha'} = R_{\alpha\alpha'} H^\alpha$, it supports a continuous infinity of non-equivalent tensor-product structures parametrised by $\alpha$. Due to the non-locality of $R_{\alpha\alpha'}$ the unitarily equivalent spaces $H^\alpha = H^\alpha_m \otimes H^\alpha_l$ and $H^{\alpha'} = H^\alpha_m \otimes H^{\alpha'}_l$ are not locally equivalent.

A. Bare-energy conservation and $\alpha$-independent predictions

In QED material systems are often interpreted as surrounded by a cloud of virtual photons [34, 40, 61–64]. Two examples of virtual processes are those described by the number non-conserving terms $ab$ and $a^\dagger b^\dagger$ having a coupling strength $u_\alpha^+$ in Eq. (3). The $\alpha$-independence of predictions is intimately related to the conservation of the bare-energy $H_0$, which determines whether a process is “real” or “virtual”. The $S$-matrix is gauge-invariant precisely because it describes “real” processes that conserve $H_0$ [47]. By contrast, virtual emission and absorption events do not individually conserve $H_0$. In this context it is noteworthy that the number non-conserving terms in Eq. (3) are $\alpha$-dependent, whereas the remaining number-conserving part is $\alpha$-independent at resonance ($\delta = 1$), which is also precisely when the bare energy is conserved; $[H_0, u_\alpha^+(ab^\dagger - a^\dagger b)] = 0$. It follows that different subsystem divisions only differ in their description of virtual processes that do not conserve $H_0$; interactions that conserve $H_0$ are $\alpha$-independent. Thus, in the traditional case of weakly-coupled nearly-resonant systems, gauge-invariance of predictions can be encoded directly at the Hamiltonian level, via the approximation of retaining only the bare-energy conserving interaction term $i\rho_b(ab^\dagger - a^\dagger b)/2$ in Eq. (3).
The idea of separating off the virtual photon cloud as a component of the electromagnetic field that is permanently tied to the material system was considered early on within the so called Pauli-Fierz representation [40]. Much more recently, the subject of ground state virtual photons has received renewed interest in strong-coupling regimes in the context of electro luminescent [66], and lossy [67] systems. There are also early experimental proposals for measuring virtual photons using solid state systems [68]. Theoretically, a representation in which ground state virtual photons are eliminated can be realized as a special case of the $\alpha$-gauge framework [35–37, 39]. In Eq. (3) this is achieved by choosing $\alpha = \alpha_{\tau} \equiv 1/(1 + \delta)$, which implies $u_{\alpha}^\tau = 0$. The interaction Hamiltonian linear in $\eta$ then has number-conserving form. The components quadratic in $\eta$ can be removed by local Bogoliubov transformations (see Supplementary Note V B). In this representation the ground state has the form of a bare vacuum, meaning that the virtual photons have been included implicitly within the so called Pauli-Fierz representation [40].

In cavity QED [28, 69], and if for example, the atom is outside the cavity when measurements are made then the two systems are obviously divisible. Furthermore, theoretically, the subsystems are uniquely defined if the coupling vanishes.

Since it is seldom tractable to include as additional quantum systems, all sources and sinks of energy that mediate the transient interaction, the most widespread method of description is to use a time-dependent coupling parameter $e(t)$, such that $e(t) = 0$ unless $t$ falls within a finite interaction period $(t_0, t_0 + \tau)$. A key point identified here is that the Hamiltonians $H^\alpha(t)$ and $H^\alpha'(t)$ obtained by replacing $e$ with a time-dependent coupling $e(t)$, in $H^\alpha$ and $H^\alpha'$ respectively, do not generate equivalent dynamics. To prove this we note that an equivalent Hamiltonian to $H^\alpha(t)$ is given by

$$
\tilde{H}^\alpha'(t) = R_{\alpha\alpha'}(t)H^\alpha(t)R_{\alpha'\alpha}(t) + i\tilde{R}_{\alpha\alpha'}(t)R_{\alpha'\alpha}(t)
$$

where $R_{\alpha\alpha'}(t)$ is obtained by making the replacement $e \rightarrow e(t)$ within the unitary gauge-fixing transformation in Eq. (4). Due to the second term on the right-hand-side of Eq. (5) the Hamiltonian $H^\alpha'(t)$, which is equivalent to $H^\alpha(t)$, is not equivalent to $H^\alpha'(t)$ unless $\alpha = \alpha'$.

- Eq. (5) implies that in the absence of an argument to fix one gauge over another when making the replacement $e \rightarrow e(t)$, the widespread method of describing tuneable light-matter interactions using an explicitly time-dependent coupling function is essentially ambiguous.

On the other hand Eq. (5) also allows us to understand why the $S$-matrix is gauge-invariant, despite the gauge-relativity of the quantum subsystems. For infinitely slow adiabatic interaction switching definitive of a scattering process, $R_{\alpha\alpha'}(t)$ in Eq. (5) is vanishingly small, so that $H^\alpha(t)$ and $H^\alpha'(t)$ are then equivalent. The gauge-invariance of the $S$-matrix can also be directly attributed to the condition of bare-energy conservation that results from its definition, as noted in the previous section [47].

In sufficiently strong-coupling non-adiabatic regimes, differences between predictions found using different gauges will occur, and may not be negligibly small. This point is especially transparent within Eq. (3), which clearly shows that the form of the interaction $V^\alpha(t)$ can be quite different depending on the value of $\alpha$ chosen. A model corresponding to $\alpha = 0$ or $\alpha = 1$, which are both commonly chosen gauges in light-matter theory, will not generally produce even qualitatively accurate predictions if the underlying physics of the system is more correctly described by an interaction corresponding, for example, to $\alpha \sim \alpha_{\tau} \equiv 1/(1 + \delta)$. In fact, we will show below that even conventional gauges $\alpha = 0$ and $\alpha = 1$, generally give significantly different physical predictions when the coupling $\eta(t)$ is ultrastrong and ultra-fast, because the two models possess different dependences on the underlying model parameters.

### III. TIME-DEPENDENT COUPLING

Subsystem properties may be of limited relevance during the light-matter interaction period. For example, concerning the interaction of a single atom with the quantized vacuum field Colen-Tannoudji et al. [34] have remarked that one cannot really remove the interaction of the charges with the transverse field and observe the bare material ground state. The bare material eigenstates and energies are not therefore operationally meaningful. They are different approximations of the real state, involving the neglect of this or that part of the effects of the transverse field on the system of charges [34]. Moreover, a strict division between light and matter systems seems increasingly artificial with increasing coupling strength.

On the other hand situations in which light and matter systems are non-interacting when they are prepared and measured, are of practical relevance for various applications. In quantum gate implementations the light and matter systems interact transiently for a finite time only. Such non-adiabatic tuneable couplings are common...
Uniform motion in and out of a cavity

In supplementary material V A we derive the Hamiltonian for atomic motion in and out of a Fabry-Perot Gaussian cavity mode with mirrors orthogonal to the z-direction, as depicted in Fig. 1. The Hamiltonian reads

\[ H^\alpha(t) = \frac{1}{2m} \left[ p + e(1 - \alpha)A(R(t)) \right]^2 + \frac{m\omega_m^2}{2} r^2 + e^2 \frac{\alpha^2 r^2}{2\nu} |\phi(R(t))|^2 - e\alpha \Pi(R(t)) \cdot r + \omega \left( a^\dagger a + \frac{1}{2} \right), \] (6)

where \( \phi(x) = e^{ikz}e^{-(x^2+y^2)/w_c^2} \) is a Gaussian mode envelope, with \( w_c \) the Gaussian beam waist, and where \( R(t) \) denotes the atom's path through the cavity. The cavity canonical operators are given by

\[ A(t, x) = \frac{1}{\sqrt{2\omega v}} \left[ \phi^*(x)a^\dagger(t) + \phi(x)a(t) \right], \] (7)

\[ \Pi(t, x) = i\sqrt{\frac{\omega}{2v}} \left[ \phi^*(x)a^\dagger(t) - \phi(x)a(t) \right]. \] (8)

Quite generally paths satisfying the condition \( \dot{z} \cdot R(t) = 0 \) have the property that the Hamiltonian in Eq. (6) is identical to that in Eq. (3) if the time-dependent coupling function in Eq. (3) is taken as \( e(t) = e\phi(R(t)) \). This confirms that the correct Hamiltonian is obtained by making the replacement \( e \to e(t) \) such that \( \eta \to \eta(t) \) in Eq. (3). Uniform motion of the dipole in and out of the cavity is described by a Gaussian function \( e(t) \). Significant \( \alpha \)-dependence of final predictions occurs when the interaction time \( \tau \sim w_c/\nu \) is comparable to the cycle time \( 1/\omega_m \). In the case of a micro-cavity with \( w_c = 20\mu m \) and \( \omega_m \) in the microwave regime, this requires \( \nu \sim 10^{-3}c \), which although relatively large, is within the non-relativistic regime.

We assume that the system starts in the ground state \( |0, 0\rangle \) and that the interaction is switched on at some time \( t_0 \) later. In Fig. 2 the number of cavity photons is plotted as a function of time with \( \eta \) chosen in the ultrastrong coupling regime and with \( w_c, \omega_m/\nu \sim 1 \), for the \( \alpha = 0, 1, \) and \( \alpha_g \) gauges. The three gauges give different residual photon populations within the cavity after the interaction has ceased. Both the Coulomb and multipolar gauges yield non-zero values. For longer interaction durations all photon populations return to zero independent of \( \alpha \).

These results afford an interpretation in terms of virtual processes that occur over short time scales, and are consistent with the suggestion of the energy-time uncertainty relation. When the interaction switch-off is long compared to the timescale of virtual processes any virtual photons emitted near the beginning of the switch-off period are reabsorbed before the interaction has ceased, i.e., within the time the atom remains inside the cavity. In contrast, when the interaction switching is on the order of a bare cycle, there is a significant probability that ground state virtual photons created at the beginning of the switch-off are not reabsorbed before the interaction has ceased. They therefore detach, and remain in the cavity once the atom has left. This occurs according to both the Coulomb and multipolar gauge interactions, but not in the \( \alpha_g \)-gauge wherein ground state virtual photons are not explicit.

![FIG. 1: A cavity of length L supporting standing waves in the z-direction and a Gaussian perpendicular mode profile with waist \( w_c \) is depicted, along with a dipole \(-\epsilon r \) oscillating with frequency \( \omega_m \). At \( t = 0 \) the cavity and dipole are non-interacting. The dipole follows a trajectory \( R(t) \) through the cavity, entering the cavity at \( t_0 \) and exiting at \( t_0 + \tau \). The Hamiltonian for this system is derived in supplementary material V A and can be realised by using a time-dependent coupling \( e(t) \) in Eq. (3).](image-url)

![FIG. 2: \( \eta = 1 \) and \( \delta = 1/2 \). The average number of photons is plotted with time in units of \( t_0 = w_c/\nu \) assuming an initial state \( |0, 0\rangle \). The beam waist is \( w_c = 20\mu m \), \( \omega_m \) is chosen in the microwave regime (energy \( \sim 10\mu eV \)) and \( \nu = 10^{-3}c \), where \( c \) is the speed of light. The beam transit time is defined by \( t_0 = w_c/\nu \). The final values are given where curves become straight, and are clearly different for different \( \alpha \).](image-url)
General time-dependent coupling

More generally, a tuneable coupling function could be used to model any time-dependent interaction, such as interactions realised by addressing specific states of the atomic system [15], or laser driven material systems [40]. Switchable interactions are also commonly encountered in superconducting circuits [17]. We therefore define the general coupling function

$$e(t)/e = 1 - \frac{\tanh \left( \frac{\omega m}{2} \right) \sinh^2 \left( \frac{\tau}{2} (t - \frac{\tau}{2} - t_0) \right)}{\cosh \left( \frac{\tau}{2} (t - t_0) \right) \cosh \left( \frac{\tau}{2} (\tau + t_0 - t) \right)}. \quad (9)$$

This is a smoothed box-function with a maximum of one at $t = t_0 + \tau/2$, such that $e(t_0)/e \approx 1/2$, and $\tau$ is roughly the full-width at half maximum. The parameter $s$ controls the smoothness of the switch-on. Through tuning of parameters this general coupling function can take a variety of forms. For example, it can be made to closely resemble a Gaussian shape, as occurs for uniform atomic motion. In what follows we determine the dependence on $\alpha$ of the final light and matter properties that result from the dynamics generated by $H^\alpha(t)$, and verify that in non-adiabatic strong-coupling regimes qualitative physical predictions do depend significantly on $\alpha$, due to the action of promoting the coupling to a time-dependent function.

A naive example of time-dependent coupling comprises instantaneous switching of a constant interaction. However, in this case the free evolution before and after the interaction window does not alter the physical quantities of interest, which makes this situation equivalent to simply considering a constant interaction $e(t) = e$. Predictions for this case in the ground state $|G\rangle$ of the full Hamiltonian $H^\alpha$ are presented in supplementary material VB. A more realistic interaction switching is smooth and therefore requires a finite time. To study this situation we use the general coupling function given in Eq. (9).

The dynamics of the system are found by numerically solving the closed set of differential equations for correlations of the form $\langle x y \rangle$ where $x, y = a, a^\dagger, b, b^\dagger$. For an initial Gaussian state of the system these correlations suffice to completely characterise the final state [70]. We find that significant $\alpha$-dependence of final predictions occurs if the switching time of the interaction is ultra-fast, i.e., of the order of a bare cycle $\omega^{-1}$, $\omega_m^{-1}$, and the coupling is sufficiently strong. For switching times longer than this predictions from different gauges converge as the interaction is switched-off, such that no differences remain by the end of the protocol. Fig. 3 shows the average number of photons in the cavity as a function of time, when the switching time is roughly $4/\omega_m$ and the system starts in the ground state $|0, 0\rangle$ of $H_0 = H^\alpha(0)$. Both initially and finally there is no ambiguity in the definitions of the light and matter systems, which are uncoupled. Relevant sub-cycle, ultrastrong couplings have already been achieved in cavity QED [15].

Since the systems are initially uncoupled it is natural to assume that they are not correlated. Correlations may then build-up due to the subsequent interaction. Fig. (4) shows the final mutual information $I(\alpha)$ at a final time $t$ long after the interaction has been switched-off, is plotted as a function of $\alpha$ for various combinations of $\delta$ and $\eta$. The inset shows the coupling envelope $e(t)/e$ as a function of time. The interaction duration given by the difference in the dashed lines is $\tau = 10/\omega_m$ with $\omega_m$ chosen in the optical range. The switch on occurs at roughly $t_0 = \tau/2$ and the chosen value of $s$ gives a switching time, represented by the arrow, of roughly $4/\omega_m$. The $\alpha$-dependence of $I(\alpha)$ varies significantly depending on the regime considered. $I(\alpha)$ is symmetric about the minimum of zero at $\alpha_g = 1/(1 + \delta)$ for all $\delta$ and $\eta$. The $\alpha$-dependence tends to be more pronounced further from resonance and for stronger coupling.
FIG. 5: $\eta = 1$ and $\delta = 3$ with $\tau$, $s$, and $\omega_0$ as in Fig. 4. $\beta_c$ corresponds to room temperature while $\beta_m = 2\beta_c$. The final subsystem energy changes and net work are plotted with $\alpha$. The net work and $\Delta E_c$ are always positive, while $\Delta E_m$ becomes negative for certain $\alpha$ implying that energy has left the initially cooler system and has entered the initially hotter system. This is due to the non-zero net work input.

generate any correlations for the values of $\eta$ and $\delta$ chosen.

To exemplify the importance of our results, we show that due to the time-dependence of the interaction even the qualitative predictions for energy exchange depend strongly on $\alpha$. To this end we consider a situation where the systems are not initially isolated from their environments. We therefore consider an initial product state of two Gibbs states $\rho(0) = \rho_m^{\alpha}(\beta_m) \otimes \rho_c^{\alpha}(\beta_c)$ where $\rho_m^{\alpha}(\beta_m) = e^{-\beta_m H_m}/\text{tr}(\cdot)$, $x = m, c$ with $H_m = \omega_m(b^\dagger b + 1/2)$ and $H_c = \omega(a^\dagger a + 1/2)$. These states result if before their interaction the systems have separately weakly-coupled and equilibrated with Markovian environments at the corresponding temperatures $\beta_m^{-1}$ and $\beta_c^{-1}$. For generality we do not assume these temperatures are equal. If the subsequent light-matter interaction is relatively short on the order of $\omega_m^{-1}$ as in Figs. 4 and 5, and is also ultrastrong, then a clear separation of time and energy scales emerges, such that weak environmental interactions can be ignored over the time-scales of interest.

Using the unitarity of the dynamics it is straightforward to show that changes in the energies of the subsystems defined by $\Delta E_x = \text{tr}[\rho_x(t)H_x] - \text{tr}[\rho_x^{eq}H_x]$ with $x = m, c$, are bounded according to $\beta_m \Delta E_m + \beta_c \Delta E_c \geq I \geq 0$ [71, 72]. If the interaction is also such that there is no net input of work, i.e., $\langle \Delta H^m(t) \rangle \equiv \langle \Delta H_0 \rangle \equiv \Delta E_m + \Delta E_c = 0$, then we obtain $(\beta_m - \beta_c)\Delta E_m \geq 0$. Thus, without a net input of work, energy cannot move from the initially cooler to the initially hotter system. On the other hand if $\Delta E_m + \Delta E_c \neq 0$ then by the end of the interaction the initially cooler system may have lost energy, with an accompanying increase in energy of the initially hotter system. Alternatively, both subsystems may simply gain energy due to the non-zero net work. The final energy that has been exchanged between the systems after the protocol has finished is shown as a function of $\alpha$ in Fig. 5. Clearly different qualitative regimes can be realised by varying only the parameter $\alpha$, which controls the gauge. Moreover, this ambiguity in qualitative predictions of final properties occurs even though the subsystems are uniquely defined at both the initial and final times.

IV. CONCLUSIONS

We have studied the implications of gauge-freedom for subsystem properties in QED, when dealing with tuneable non-adiabatic, strong-coupling. When the coupling is non-vanishing there are infinitely many non-equivalent definitions of the quantum subsystems. For strong enough coupling “light” and “matter” subsystem properties like entanglement and photon number, are significantly different for different subsystem definitions. These differences persist in the case of tuneable interactions, and become increasingly pronounced as the coupling switching increases in strength and speed.

Obtaining even qualitatively reliable predictions of final physical properties is dependent on identifying the correct coupling model when describing a given physical set-up. Predictions regarding the generation and manipulation of entanglement and other subsystem properties cannot be relied upon without a concrete justification that the chosen coupling model accurately describes the physical set-up considered. This finding is of major importance for current technological applications including quantum communication, metrology, simulation, and information processing, in which final subsystem properties are of central importance. The description given here should be extendable to time-dependent interactions that arise in solid state systems, organic systems, and superconducting circuits. This will reveal the extent of the ambiguity across the platforms of current interest in the applications of quantum science.

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V. SUPPLEMENTARY MATERIAL

A. Atom moving in and out of a Fabry-Perot cavity

In this section we derive the Hamiltonian describing the interaction between a Fabry-Perot cavity with an oscillating dipole at an arbitrary position within the cavity.

1. Quantisation of the free cavity

We consider a Fabry-Perot cavity consisting of parallel mirrors in the $xy$-plane separated by a distance $L$. In the $z$-direction the electromagnetic field satisfies periodic boundary conditions, with a Gaussian profile in the perpendicular direction $x\hat{x} + y\hat{y}$ [73]. We restrict our attention to the fundamental Gaussian mode in the perpendicular direction. Although not necessary, for simplicity we also consider only the fundamental standing wave mode in the $z$-direction. One could also consider additional Gauss-Hermite or Gauss-Laguerre modes in the perpendicular direction.

In the present case the single cavity mode is described by a pure Gaussian beam propagating in the $z$-direction such that classically the transverse vector potential is

$$A(t, x) = \varepsilon A_0 u(x)e^{-i\omega t} + c.c.$$ (10)

where $\varepsilon$ is a transverse polarisation in the $xy$-plane and $u(x)e^{-i\omega t}$ satisfies the paraxial scalar wave equation [73–75]. Anticipating the transition to the quantum theory we have written the space and time-independent amplitude $A_0$ as the product of a real normalisation $A$ and a complex number $a$. We have also neglected a small non-transverse component in the $z$-direction [74, 75]. We define $\Pi(t, x) = A(t, x) \equiv -E_T(t, x)$ such that the cavity energy is

$$H_I = \frac{1}{2} \int d^3 x [E_T(x)^2 + B(x)^2] = \int d^3 x \Pi(x)^2$$ (11)

where $\int \prime$ indicates that spatial integration is restricted to the cavity length $L$ in the $z$-direction, and $B = \nabla \times A$. We have also assumed that the magnetic and electric energy-densities are the same in the free theory.

To obtain an explicit expression for $H_I$ that can be quantised we consider the fundamental Gaussian mode solution to the paraxial wave equation $u(x)e^{-i\omega t}$ such that [75]

$$u(x) = \frac{w_c}{w(z)} e^{-\left(x^2+y^2\right)/w(z)^2} e^{ik\left(x^2+y^2\right)/2R(z) + i\theta(z) + ikz}$$ (12)

with $(0, 0, k)$ the wave-vector such that $k = \omega$ and

$$z_R = \frac{1}{2} kw_c^2, \quad w(z) = w_c \sqrt{1 + \left(\frac{z}{z_R}\right)^2},$$

$$R(z) = z + \frac{z_R^2}{z}, \quad \theta(z) = -\arctan \frac{z}{z_R},$$ (13)

where $w_c$ denotes the beam waist. For $L \ll z_R$ we have $w(z) \approx w_c$, $k(x^2 + y^2)/2R(z) \approx 0$ and $\theta(z) \approx -\pi/2$. In this limit Eq. (12), reduces to

$$u(x) \approx \phi(x) = e^{ikz} e^{-\left(x^2+y^2\right)/w_c^2}$$ (14)
where we have ignored a global phase $e^{-i\pi/2}$. We define the cavity volume by

$$v = \frac{1}{2} \int d^3x |\phi(x)|^2 = \frac{\pi w^2 L}{2}$$

(15)

and choose the normalisation $\mathcal{A} = 1/\sqrt{2\omega v}$, such that substitution of Eq. (10) into the right-hand-side of Eq. (11)

yields

$$H_I = \frac{\omega}{2}(a^+a + aa^+) = \frac{v}{2}(\Pi^2 + \omega^2 A^2)$$

(16)

where $A \equiv A(x = 0)$ and $\Pi \equiv \Pi(x = 0)$. This cavity Hamiltonian is formally identical to the bare-cavity Hamiltonian of Sec. II, and in the free (non-interacting) theory it is $\alpha$-independent. In obtaining Eq. (16) we have used

$$\int d^3x \phi(x)^2 = \int_0^{l+L} dL e^{2ikz} \int dx dy e^{-(x^2+y^2)/w^2} = 0,$$

(17)

where $l$ is arbitrary such that $l$ and $l + L$ are the positions of the two cavity mirrors along the $z$-axis. Eq. (17) follows

from the vanishing of the $z$-integral due to the periodic boundary conditions in the $z$-direction; $k = n\pi/L$, $n = 0, 1, 2, 3,...$

Quantisation is now straightforward via the replacement of the complex numbers $a$ and $a^*$ with bosonic operators $a$ and $a^\dagger$ such that $[a, a^\dagger] = 1$. We thereby obtain the mode expansions

$$A(t, x) = \frac{\varepsilon}{\sqrt{2\omega v}}[\phi^*(x)a^\dagger(t) + \phi(x)a(t)],$$

(18)

$$\Pi(t, x) = i\varepsilon \sqrt{\frac{\omega}{2v}}[\phi^*(x)a^\dagger(t) - \phi(x)a(t)],$$

(19)

where $a(t) = ae^{-i\omega t}$ in the free theory. All non-zero equal-time canonical commutation relations are obtained from Eqs. (18) and (19) using $[a, a^\dagger] = 1$;

$$[A_i(t, x), A_j(t, x')] = i\frac{\varepsilon_i \varepsilon_j}{2v} [\phi(x)\phi^*(x') + \phi^*(x)\phi(x')],$$

(20)

$$[A_i(t, x), A_j(t, x')] = i\frac{\varepsilon_i \varepsilon_j}{2\omega v} [\phi(x)\phi^*(x') - \phi^*(x)\phi(x')],$$

(21)

$$[\Pi_i(t, x), \Pi_j(t, x')] = \frac{\omega}{2}[A_i(t, x), A_j(t, x')].$$

(22)

In particular we have $[A_i, \Pi_j] = i\varepsilon_i \varepsilon_j v/2$ and $[A_i, A_j] = 0 = [\Pi_i, \Pi_j]$ in agreement with Sec. II.

The violation of relativistic causality implied by the non-vanishing commutators of fields at spacelike separated events is a result of the approximations made, namely the restriction to a single radiation mode and the paraxial approximation. The single-mode approximation eliminates the spatio-temporal structure necessary to elicit causality and has been discussed in this context recently in Ref. [76]. These authors consider the propagation direction only and show that by including more standing wave modes consistency with relativistic causality is recovered. Here, our aim is to study the role of the gauge-parameter $\alpha$ in the light-matter interaction and for this purpose it suffices to restrict attention to the fundamental mode. As noted at the beginning of this section the single-mode approximation is certainly not necessary and has been used here for simplicity. Without requiring any essentially new theoretical machinery one can extend the present treatment in a straightforward manner to include more modes in the transverse direction or in the $z$-direction. Within the single-mode treatment, which is adequate for the present purpose, the canonical commutation relations (20)-(22) are necessary for the formal self-consistency of the framework developed.

2. Cavity-dipole interaction

Let us now consider the coupling of the cavity to an oscillating dipole located at an arbitrary position $R$. Expressed in the Coulomb gauge the light-matter Hamiltonian is

$$H^0 = \frac{1}{2m} |p + eA(R)|^2 + \frac{m\omega^2}{2} r^2 + H_I.$$

(23)
If we fix \( \mathbf{R} = 0 \) this Hamiltonian is identical to that given by Eq. (3) with \( \alpha = 0 \). The Coulomb gauge expression is of course only one possible way to express the Hamiltonian. We can obtain a more general expression via the gauge-transformation \( R_{\alpha \alpha'} = e^{-i e(\alpha - \alpha') \mathbf{r} \cdot \mathbf{A}(\mathbf{R})} \), which reduces to that in Eq. (4) when \( \mathbf{R} = 0 \). In particular,

\[
\begin{align*}
R_{0 \alpha} p R_{0 \alpha}^{-1} &= p - e \alpha \mathbf{A}(\mathbf{R}), \\
R_{0 \alpha} \Pi(x) R_{0 \alpha}^{-1} &= \Pi(x) - \frac{e c \mathbf{r}}{2 \omega} [\phi(\mathbf{R}) \phi^*(x) + \phi^*(\mathbf{R}) \phi(x)], \\
R_{0 \alpha} A(x) R_{0 \alpha}^{-1} &= A(x) + \frac{ie c \mathbf{r}}{2 \omega v} [\phi(\mathbf{R}) \phi^*(x) - \phi^*(\mathbf{R}) \phi(x)], \\
R_{0 \alpha} a R_{0 \alpha}^{-1} &= a - \frac{ie \alpha (\mathbf{r} \cdot \mathbf{e})}{\sqrt{2 \omega v}} \phi^*(\mathbf{R}).
\end{align*}
\]

(24)

Only the operators \( \mathbf{r} \) and \( \mathbf{A}(\mathbf{R}) \), which commute with \( R_{\alpha \alpha'} \) are \( \alpha \)-independent. The Hamiltonian in the gauge \( \alpha \) is obtained using Eqs. (24) as

\[
H^\alpha = R_{0 \alpha} H^0 R_{0 \alpha}^{-1} = \frac{1}{2m} [p + e(1 - \alpha) \mathbf{A}(\mathbf{R})]^2 + \frac{m c^2}{2} \mathbf{r}^2 + e^2 \frac{\alpha^2 \mathbf{r}^2}{2 \omega} \phi(\mathbf{R})^2 - e \alpha \mathbf{A}(\mathbf{R}) \cdot \mathbf{r} + \omega \left( a^\dagger a + \frac{1}{2} \right),
\]

(25)

which reduces to that given by Eq. (3) if \( \mathbf{R} = 0 \). The framework developed in this section therefore generalises that of Sec. II by providing explicit mode expansions for the cavity canonical operators in the form of Eqs. (18) and (19), which can be evaluated at an arbitrary atomic position \( \mathbf{R} \). As before \( \alpha \) controls the gauge with \( \alpha = 0 \) and \( \alpha = 1 \) yielding the Coulomb and multipolar gauges respectively.

If the dipole is moving its position \( \mathbf{R} \) is time-dependent \( \mathbf{R} = \mathbf{R}(t) \). Without loss of generality we can consider cavity mirrors located at \( z = \pm L/2 \) centred at \( (0,0) \) in the \( xy \)-plane. Any prescribed dipolar motion may now be considered. The simplest case consists of uniform motion \( -\mathbf{v} \mathbf{X} \) starting from rest at the point \( \mathbf{h} \mathbf{X} \), which yields the path \( \mathbf{R}(t) = \mathbf{h}(h - vt) \). A second example consists of the dipole dropped from \( \mathbf{h} \mathbf{X}/2 \) falling under gravity with trajectory \( \mathbf{R}(t) = (h - gt^2/2) \mathbf{X} \) towards the ground at \( -\mathbf{h} \mathbf{X}/2 \). Here \( g \) is the magnitude of acceleration due to gravity, assumed to be uniform. Both of these examples give paths such that \( \mathbf{z} \cdot \mathbf{R}(t) = 0 \) for all \( t \). Quite generally paths satisfying this condition have the property that the Hamiltonian in Eq. (25) with \( \mathbf{R} = \mathbf{R}(t) \) is identical to that in Eq. (3) if the time-dependent coupling function in Eq. (3) is taken as \( e(t) = e \phi(\mathbf{R}(t)) \). For the two examples described above the coupling functions are \( e(t)/e = e^{-(h - vt)^2/w_n^2} \) in the case of uniform motion and \( e(t)/e = e^{-(h - gt^2/2)^2/w_n^2} \) in the case of motion under gravity. Thus, we see that the replacement \( e \to e(t) \) is a well-justified treatment of the time-dependent interaction resulting from a dipole with prescribed gross motion.

\[ \text{FIG. 6: The mutual information } I(\alpha, t) \text{ is plotted with time in units of } t_b = w_n/\nu \text{ assuming an initial state } |0,0\rangle. \text{ All parameters are as in Fig. 2.} \]

In the case of uniform motion the Gaussian coupling envelope incurs a relatively smooth switch-on. For a beam waist \( w_n = 20 \mu m \), with \( h \) substantially larger, so that the dipole starts well outside of the cavity, and for a dipole with microwave frequency \( \omega_m \sim \text{GHz} \), the gross dipolar speed must be around \( \nu = 10^{-3} c \) in order that the interaction time \( \tau \sim w_n/\nu \) is comparable to the cycle time \( 1/\omega_m \). The velocity \( 10^{-3} c \) is not yet relativistic, but significantly larger than the velocities found in typical atomic beam experiments, which are around three orders of magnitude smaller.
In order to achieve $w_c\omega_m/\nu \sim 1$ with smaller $\nu$ either the cavity beam waist must be further reduced, or slower dipolar oscillations must be considered. However $w_c\omega_m/\nu \sim 1$ is achieved, significant differences occur in predictions associated with different gauges within this regime, as shown in Fig. 6.

B. Ground state of the interacting Hamiltonian, and the ground state photon number and mutual information

A naive example of a time-dependent interaction comprises instantaneous interaction switch-on/off described by the function $e(t) = e[u(t-t_0) - u(t-(t_0+\tau))]$ where $u$ denotes the unit-step function. For final times $t > t_0 + \tau$ the evolution of the system is composed of sequential evolutions as $U(0, t) = U_0(0, t_0)U^\alpha(t_0, t_0 + \tau)U_0(t_0 + \tau, t)$ where $U^\alpha(t', t) = e^{-i(t-t')}H^\alpha$ and $U_0(t', t) = e^{-i(t-t')}H_0$. However, the free (uncoupled) evolution $U_0$ does not alter either the oscillator populations nor the final light-matter correlations. To find these observables one can set $t_0 = 0$ and $\tau = t$ without loss of generality, which is equivalent to considering the full interacting system with a constant interaction $e(t) = e$. In this case it is more physically relevant to consider an initial eigenstate of the full Hamiltonian $H^\alpha$ rather than the free part $H_0$.

Of considerable interest are light-matter correlations in the ground state $|G\rangle$ of the full Hamiltonian $H^\alpha$. These are quantified by the mutual information $I_G(\alpha) = S(\rho_m^\alpha) + S(\rho_l^\alpha)$ where $S(\rho) = -\text{tr}\rho \ln \rho$ and the reduced material and cavity states are defined by $\rho_m^\alpha = \text{tr}_l |G\rangle \langle G|$ and $\rho_l^\alpha = \text{tr}_m |G\rangle \langle G|$ respectively. The mutual information $I_G(\alpha)$ is found to be

$$I_G(\alpha) = (\mu_\alpha + 1) \ln \left( \frac{\mu_\alpha + 1}{2} \right) - (\mu_\alpha - 1) \ln \left( \frac{\mu_\alpha - 1}{2} \right)$$

(26)

where

$$\mu_\alpha = \sqrt{1 + \left( \frac{\omega}{\omega_g} \right)^2 \frac{e^2}{mv\omega_m} (\alpha - \alpha_g)^2}.$$

(27)

It is symmetric about the point $\alpha = \alpha_g$ where it takes its minimum value of zero.

We also consider the average number of $\alpha$-gauge photons $n_\alpha(\alpha) = \langle a^\dagger a \rangle_G$ in the ground state. A straightforward calculation yields

$$n_\alpha(\alpha) = \frac{1}{4\omega} \left[ \omega_g + \frac{e^2(\alpha - \alpha_g)^2}{mv\omega_m g} + \frac{\omega^2}{\omega_g} - \frac{1}{2} \right]$$

(28)

where $\omega_g \equiv \omega_{\alpha_g}$ and

$$\alpha_g = \frac{\omega_m}{\omega_m + \omega}, \quad \omega_{m,g}^2 = \omega_m^2 + \frac{e^2}{mv}\alpha_g^2.$$

(29)

If one allows the definition of photon number to depend on material parameters $e$ and $m$ then the self-energy term

$$e^2(1 - \alpha)^2 \alpha A^2 / 2m = \eta^2 \omega [(1 - \alpha)^2 (a^\dagger a)^2]$$

(30)
can be absorbed into a redefinition of the local cavity energy as

$$\hat{H}_l^\alpha = H_l^\alpha + e^2(1 - \alpha)^2 \alpha A^2 / 2m = \frac{\nu}{2} (\Pi + \omega_\alpha^2 \alpha^2) = \omega_\alpha \left( c^\dagger c + \frac{1}{2} \right)$$

(31)

where

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

(32)

The operators $c$, $c^\dagger$ are related to $a$, $a^\dagger$ by a local Bogoliubov transformation in $H_l^\alpha$. The average number of $\alpha$-gauge renormalised ground state photons $n_\alpha(\alpha) = \langle c^\dagger c \rangle_G$ is

$$n_\alpha(\alpha) = \frac{1}{4\omega_\alpha} \left[ \omega_g + \frac{e^2(\alpha - \alpha_g)^2}{mv\omega_m g} + \frac{\omega^2}{\omega_g} - \frac{1}{2} \right].$$

(33)
\[ I_G(\alpha) \text{ is plotted as a function of } \alpha \text{ with } \delta = \omega/\omega_m = 1/2, \text{ for three values of the dimensionless coupling parameter } \eta = e/(\sqrt{\omega m}). \text{ The strength of the } \alpha\text{-dependence increases with increasing } \eta. \text{ For all } \eta \text{ the mutual information } I_G(\alpha) \text{ is symmetric about the minimum value of zero occurring at } \alpha_g = 1/(1 + \delta) \text{ for which the ground state } |G\rangle \text{ is in fact separable (supplementary material V B). At resonance } \delta = 1 \text{ we have } \alpha_g = 1/2, \text{ implying } I_G(0) = I_G(1). \text{ Off-resonant values of } \delta \text{ determine the shift of the minimum } \alpha_g \text{ relative to the resonant value; } \alpha_g \text{ is shifted towards } \alpha = 1 \text{ for } \delta < 1, \text{ and towards } \alpha = 0 \text{ for } \delta > 1.\]

Unlike the average in Eq. (28) this average reaches a minimum of zero for } \alpha = \alpha_g. \text{ This can be understood by noting that for this choice of } \alpha \text{ the Hamiltonian can be written in number-conserving from as}

\[ H^g = \omega_{m,g} \left( d^\dagger d + \frac{1}{2} \right) + \omega_g \left( c^\dagger c + \frac{1}{2} \right) + ie \sqrt{\frac{\omega_m}{m\nu}} \frac{1}{\omega_m + \omega} (d^\dagger c - dc^\dagger) \]

(34)

where the renormalised material modes } d \text{ are such that}

\[ \frac{p^2}{2m} + \frac{m\omega_{m,g}^2}{2} r^2 = \omega_{m,g} \left( d^\dagger d + \frac{1}{2} \right). \]

(35)

The renormalised material modes } d \text{ are connected to the bare material modes } b \text{ via a local Bogoliubov transformation in } H^g_{cm}. \text{ The ground state } |G\rangle \text{ of the Hamiltonian is the vacuum } |0^d, 0^c\rangle \text{ annihilated by the operators } d \text{ and } c. \text{ Thus, } n_d(\alpha_g) = 0. \text{ It is important to note that unlike a full diagonalisation of the Hamiltonian, the partially diagonal form Eq. (34) does not obscure the divisibility of the overall system into “light” and “matter” subsystems. After a full diagonalisation the Hamiltonian can be written as the sum of two harmonic oscillator energies, but it is not possible to distinguish these harmonic oscillators such that one can be called “light” and the other “matter” in any meaningful way. This is because a completely diagonalising transformation is necessarily non-local with respect to the light-matter Hilbert space bipartition of any gauge. On the other hand the number-conserving form Eq. (34) can be achieved by simply choosing a particular gauge and then performing nothing but local operations within that gauge.}

Fig. 7 shows significant variations in the mutual information } I_G(\alpha), \text{ which become increasingly pronounced for larger dimensionless coupling-strengths } \eta. \text{ Similarly Fig. 8 plots } n_d(\alpha) \text{ and } n_c(\alpha) \text{ as a functions of } \alpha, \text{ showing that both non-renormalised and renormalised photon numbers vary significantly with } \alpha.
FIG. 8: The ground state photon number averages $n_a(\alpha)$ and $n_c(\alpha)$ are plotted as functions of $\alpha$ with $\delta = \omega/\omega_m = 2$. The strength of the dependence on $\alpha$ increases with increasing $\eta$, as does the difference between the two photon numbers $n_a$ and $n_c$. For sufficiently weak coupling $\eta \leq 0.1$, $n_a$ and $n_c$ are indistinguishable within the resolution of the plot. Both $n_a$ and $n_c$ are minimum at $\alpha = \alpha_g$. For all couplings $n_c$ is identically zero at $\alpha_g$ while $n_a$ becomes non-zero for stronger coupling. For $\alpha \to 1$, $n_a(\alpha) \to n_c(\alpha)$, because the self-energy term $e^2(1 - \alpha)^2 A^2/2m$ vanishes identically in the Poincaré gauge $\alpha = 1$. 