Supplemental Material for "Spontaneous Scattering of Raman Photons from Cavity-QED Systems in the Ultrastrong Coupling Regime"

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I. HAMILTONIAN IN THE DIPOLE GAUGE

In the dipole gauge, the field conjugate momentum corresponds to the displacement operator $\hat{\mathbf{\Pi}}(\mathbf{r}) = -\hat{\mathbf{D}}(\mathbf{r}) = -i\epsilon_0 \mathbf{E}_0(\mathbf{r})(\hat{a}-\hat{a}^{\dagger})$, where $\mathbf{E}_0(\mathbf{r}) = \sqrt{\frac{\omega_c}{2\epsilon_0}} \mathbf{f}_0(\mathbf{r})$, with $\mathbf{f}_0(\mathbf{r})$ the mode function of the cavity mode under consideration. In this gauge, the electric field operator is not described only by photon operators; taking into account that $\epsilon_0 \hat{\mathbf{E}} = \hat{\mathbf{D}} - \hat{\mathbf{P}}$, respectively, one can prove [1] that $\hat{\mathbf{E}}$ adopts the form $\hat{\mathbf{E}} \approx \mathbf{E}_0(\mathbf{r})[i(\hat{a}-\hat{a}^{\dagger})-2\eta\hat{\sigma}_p]$, where $\eta \equiv \boldsymbol{\mu} \cdot \mathbf{E}_0(\mathbf{r})/\omega_c$ is the dimensionless coupling parameter between cavity and TLS, and we have neglected the small contribution of the polarizability of the ancilla sensor. In the dipole gauge, the interaction Hamiltonian thus takes the form [1, 2] ($\hbar = 1$),

$$\hat{H}_I = i\eta\omega_c(\hat{a}^{\dagger} - \hat{a})\hat{\sigma}_p + \omega_c\eta_s[i(\hat{a}^{\dagger} - \hat{a}) + 2\eta\hat{\sigma}_p]\hat{\sigma}_x^s.$$
(S1)

This Hamiltonian ensures a proper gauge invariance under the TLS approximation [2–4]. Here, $\eta_s \equiv \mu_s \cdot \mathbf{E}_0(\mathbf{r}_0)/\omega_c$ is the coupling parameter between sensor and cavity.

In this work we will focus on phenomenology emerging on the ultrastrong coupling regime, where $\eta \sim 0.1$, and fix $\eta_s \ll 1$ so that the sensor qubit only probes the dynamics of the TLS-cavity system, without altering it. We observe that the sensor-atom coupling term in Eq. (S1) origins from the matter self-interaction term which is present in the dipole gauge and ensures gauge invariance.

From now on, we consider that the problem is confined to a single polarization, i.e., all vector quantities are aligned along the same unit vector \mathbf{u}_x , and substitute vector field operators $\hat{\mathbf{E}}$ by scalar operators, so that $\hat{\mathbf{E}} = \hat{E}\mathbf{u}_x$. Finally, the drive Hamiltonian reads $\hat{H}_{\text{drive}}(t) = \Omega[i(\hat{a} - \hat{a}^{\dagger}) - 2\eta\hat{\sigma}_x]\cos(\omega_{\text{L}}t)$, so that $\hat{H}_{\text{drive}} \propto \hat{E}\cos(\omega_{\text{L}}t)$.

II. DRESSED OPERATORS: PHOTON DETECTION AND DISSIPATION IN THE DIPOLE GAUGE

When the normalized coupling strength become significantly large, the peculiar relevance of the counter-rotating light-matter interaction terms has dramatic consequences on nearly all aspects of the theory, both in open and closed systems. As a result,

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the ground-state and all excited-states are dressed and contain different number of virtual bare excitations [5, 6]. This implies that the treatment of [7-11] dissipation, input-output relationships, correlations, driving, and photon detection rate is not the trivial extension of standard quantum optics procedures, but requires to correctly take into account that virtual excitations cannot escape the system.

In doing this, one has to introduce the so called dressed basis $\{|j\rangle\}$, obtained by diagonalizing the undriven system Hamiltonian [see eq. (1) in the main text] $\hat{\mathcal{H}} = \hat{H}_{\text{free}} + \hat{H}_{\text{I}} = \sum_{j} \omega_{j} |j\rangle \langle j|$. Each operators will be rotate with respect to the dressed basis and then separate in its positive and negative frequency components obtaining two new operators able to describe correctly the ultrastrong coupling physics,

$$\hat{\mathcal{O}}^{+} = \sum_{j=1}^{N} \sum_{k>j} \langle j | \hat{\mathcal{O}} | k \rangle | j \rangle \langle k |$$

$$\hat{\mathcal{O}}^{-} = [\hat{\mathcal{O}}^{+}]^{\dagger},$$
(S2)

where k > j for $\omega_k > \omega_j$.

The emitted-photon rate (at given time), which can be measured by a broadband point-like detector in the resonator [10], is proportional to the expectation value $\langle \hat{\mathcal{X}}^- \hat{\mathcal{X}}^+ \rangle_t \equiv \text{Tr}[\hat{\mathcal{X}}^- \hat{\mathcal{X}}^+ \hat{\rho}(t)]$, where $\hat{\rho}(t)$ is the density operator of the light-matter system. For cavity resonators ultrastrongly coupled to a two-level system $\hat{\mathcal{X}}^+$ is the positive-frequency electric field operator in the dipole gauge, which destroys a physical cavity excitation,

$$\hat{\mathcal{X}}^{+} = \sum_{j=1}^{k} \sum_{k>j} \langle j | [i\omega_{c}(\hat{a} - \hat{a}^{\dagger}) - 2\omega_{c}\eta\hat{\sigma}_{x}] | k \rangle | j \rangle \langle k |, \qquad (S3)$$
$$\hat{\mathcal{X}}^{-} = [\hat{\mathcal{X}}^{+}]^{\dagger}.$$

By using the input-output theory, analogous results can be obtained for the output-rate of emitted photons detected by a detector placed outside the cavity [8]. However, the output field operators can display a different dependence on $\omega_{kj} = \omega_k - \omega_j$, arising from the output-modes density of states and from the frequency dependence of the coupling coefficient (which depends on the mirror reflectivity in a standard microcavity). The output field operator in the dipole gauge, encoding into the model-dependent function $\alpha(\omega)$ such a dependence, reads as,

$$\hat{\mathcal{E}}_{out}^{+} = i \sum_{j=1}^{N} \sum_{k>j} \alpha(\omega_{kj}) \langle j | (\hat{a} + \hat{a}^{\dagger}) | k \rangle | j \rangle \langle k | \,.$$
(S4)

Starting from the relation $\hat{D} = -\hat{A}$ and using the generalized TRK sum rule for electromagnetic fields [12] (which is valid even in the presence of very strong light-matter interactions and/or optical nonlinearities), it is possible to demonstrate that,

$$\omega_c \langle j | (\hat{a} - \hat{a}^{\dagger} + i2\eta \hat{\sigma}_x) | k \rangle = \omega_{kj} \langle j | \hat{a} + \hat{a}^{\dagger} | k \rangle.$$
(S5)

Therefore, if we assume $\alpha_c(\omega_{kj}) = \omega_{kj}$ to be linearly dependent on the transition frequencies, one gets $\hat{\mathcal{E}}_{out}^+ = \hat{\mathcal{X}}^+$. A different choice will give rise to similar spectra with different relative heights of the spectral lines. Notice that photodetection is an energy absorption process, thus it is reasonable to assume photodetection rates which tend to zero with frequencies $\omega \to 0$.

Analogously, it is possible to define the field operators describing the qubit emission $\sim \langle \hat{\Sigma}^- \hat{\Sigma}^+ \rangle_t \equiv \text{Tr}[\hat{\Sigma}^- \hat{\Sigma}^+ \hat{\rho}(t)]$. By using input-output theory, in a similar manner one can get same result for the qubit part,

$$\hat{\Sigma}_{out}^{+} = \hat{\Sigma}^{+} = i \sum_{j=1}^{\infty} \sum_{k>j} \frac{\omega_{kj}}{\omega_q} \langle j | \hat{\sigma}_x | k \rangle | j \rangle \langle k |$$

$$\hat{\Sigma}^{-} = [\hat{\Sigma}^{+}]^{\dagger}.$$
(S6)

We observe that $\hat{\sigma}_x$ is the only gauge-invariant Pauli matrix.

The generalized master equation (GME) [9] for strongly interacting hybrid quantum systems is developed using the dressed basis of the undriven system Hamiltonian, without applying post-trace approximation. The latter assumption makes the dissipation processes suitable for systems which display also harmonic or quasi-harmonic spectra. It goes beyond the dressed master equation in Ref. [7], suitable only for anharmonic systems. This feature is essential to describe the system dynamics and the resulting spectra [13] both at small and at very large (deep strong) values of the normalized coupling strength η . The GME in the dipole gauge for the Rabi model can be written as,

$$\dot{\hat{\rho}} = -i \left[\hat{\mathcal{H}}_d, \hat{\rho} \right] + \mathcal{L} \hat{\rho} \,. \tag{S7}$$

The Liouvillian superoperator \mathcal{L} contains two contributions $\mathcal{L} = \mathcal{L}^{c} + \mathcal{L}^{q}$, arising from the cavity-bath and the qubit-bath interaction,

$$\mathcal{L}^{c(q)}[\hat{\rho}] = \frac{1}{2} \sum_{\substack{j > k \\ l > m}} \{ \Gamma_{lm}^{c(q)} n_{ml}(\mathcal{T}_{c(q)}) [\hat{P}_{lm} \hat{\rho} \hat{P}_{kj} - \hat{P}_{kj} \hat{P}_{lm} \hat{\rho}] \\ + \Gamma_{kj}^{c(q)} n_{jk}(\mathcal{T}_{c(q)}) [\hat{P}_{lm} \hat{\rho} \hat{P}_{kj} - \hat{\rho} \hat{P}_{kj} \hat{P}_{lm}] \\ + \Gamma_{lm}^{c(q)} [n_{ml}(\mathcal{T}_{c(q)}) + 1] [\hat{P}_{kj} \hat{\rho} \hat{P}_{lm} - \hat{\rho} \hat{P}_{lm} \hat{P}_{kj}] \\ + \Gamma_{kj}^{c(q)} [n_{jk}(\mathcal{T}_{c(q)}) + 1] [\hat{P}_{kj} \hat{\rho} \hat{P}_{lm} - \hat{P}_{lm} \hat{P}_{kj} \hat{\rho}] \}.$$
(S8)

The $\hat{P}_{kj} = |k\rangle\langle j|$ are the projection operators, and $n_{kj}(\mathcal{T}_{c(q)}) = [\exp(\omega_{kj}/(\omega_{c(q)}\mathcal{T}_{c(q)})) - 1]^{-1}$ are the thermal populations of the cavity (c) and qubit (q) reservoirs calculated at the transition frequencies. The cavity-bath and qubit-bath coupling rates determine both the losses and the incoherent pumping of the system, depending on the specific interaction Hamiltonian and on the baths density of states. From the general derivation and using Eq. (S5) one obtains,

$$\Gamma_{kj}^{c} = \kappa \frac{\omega_{kj}}{\omega_{c}} |\langle j|(\hat{a} + \hat{a}^{\dagger})|k\rangle|^{2}, \qquad \Gamma_{kj}^{q} = \gamma \frac{\omega_{kj}}{\omega_{q}} |\langle j|\hat{\sigma}_{x}|k\rangle|^{2}.$$
(S9)

For systems interacting with an environment at temperature $\mathcal{T} = 0$, the Liouvillian superoperator \mathcal{L} gets a simpler form which can be expressed in terms of dressed operators $\hat{\mathcal{X}}^{+(-)}$ in Eq. (S3), and $\hat{\Sigma}^{+(-)}$ in Eq. (S6) being the cavity and qubit dissipator

$$\mathcal{L}_{\hat{\mathcal{X}}^{+}}[\hat{\rho}] = \hat{\mathcal{X}}^{+}\hat{\rho}\hat{\mathcal{X}}^{-} - \frac{1}{2}\{\hat{\mathcal{X}}^{-}\hat{\mathcal{X}}^{+},\hat{\rho}\},\$$

$$\mathcal{L}_{\hat{\Sigma}^{+}}[\hat{\rho}] = \hat{\Sigma}^{+}\hat{\rho}\hat{\Sigma}^{-} - \frac{1}{2}\{\hat{\Sigma}^{-}\hat{\Sigma}^{+},\hat{\rho}\}.$$
 (S10)

The presence of an auxiliary qubit sensor $\hat{\sigma}_z^s$ of frequency ω_s coupled to the cavity mode, makes a new dissipator necessary. The dissipator for the qubit sensor can be constructed in the same manner, following our previous discussion, as

$$\mathcal{L}_{\hat{\Sigma}_{s}^{+}}[\hat{\rho}] = \hat{\Sigma}_{s}^{+} \hat{\rho} \hat{\Sigma}_{s}^{-} - \frac{1}{2} \{ \hat{\Sigma}_{s}^{-} \hat{\Sigma}_{s}^{+}, \hat{\rho} \}.$$
(S11)

III. AVERAGED STATIONARY STATE OF A TIME-DEPENDENT PROBLEM: FLOQUET THEORY

Due to the presence of counter-rotating terms in the Hamiltonian

$$\hat{H} = i\eta\omega_c(\hat{a}^{\dagger} - \hat{a})\hat{\sigma}_p + \omega_c\eta_s[i(\hat{a}^{\dagger} - \hat{a}) + 2\eta\hat{\sigma}_p]\hat{\sigma}_x^s + \Omega[i(\hat{a} - \hat{a}^{\dagger}) - 2\eta\hat{\sigma}_x]\cos(\omega_{\rm L}t),$$
(S12)

one cannot perform the standard unitary transformation to the rotating frame of the drive that leads to a time-independent Hamiltonian, and thus we are left with the explicitly time-dependent terms. The Liouvillian superoperator (defined as the generator of the master equation $\dot{\rho} = \mathcal{L}\rho$) can then be cast as $\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_+ e^{i\omega_{\rm L}t} + \mathcal{L}_- e^{-i\omega_{\rm L}t}$. As a consequence, there is not a fully stationary state in the long-time limit, but a time-dependent state whose form we can postulate as $\rho(t \to \infty) = \sum_{n=-\infty}^{\infty} \rho_n e^{in\omega_{\rm L}t}$, which allows us to solve the problem using Floquet theory [14–16]. The time-averaged steady-state matrix is $\rho^{\rm ss} = \rho_0$, and can be found as the nullspace of $\mathcal{L}_0 + \mathcal{L}_- \mathcal{S}_1 + \mathcal{L}_+ \mathcal{S}_{-1}$, where the \mathcal{S}_n operators are obtained recursively as

$$\mathcal{S}_{\pm n} = -[\mathcal{L}_0 - (z + in\omega_{\rm L}) + \mathcal{L}_{\mp}\mathcal{S}_{\pm(n+1)}]^{-1}\mathcal{L}_{\pm},\tag{S13}$$

which is solved assuming that $S_{\pm n_{\max}} = 0$ for a sufficiently large n_{\max} .

Once a time-averaged steady state is obtained, we consider that the stationary rate of emission from the sensor is proportional to the spectrum of emission at the sensor's frequency, i.e. $S(\omega_s) \propto \text{Tr}[\rho_{ss}\hat{\Sigma}_s^-\hat{\Sigma}_s^+]$, with the sensor's decay rate Γ corresponding to the filter linewidth. One is thus able to compute the spectrum by scanning the sensor's frequency and computing the corresponding time-averaged steady state.

IV. IMPLICATIONS OF RAMAN PEAKS FOR SPECTROSCOPY: FISHER INFORMATION

The identification of methods to improve the characterization of cavity QED systems is an important task, given that these are an integral part of many architectures in quantum technologies. The description and understanding of Raman scattering



FIG. S1. Estimation of cavity QED parameters with Raman peaks. Spectrum around the frequency of the Stokes transition $\omega_{\rm S}^{(10)}$ (a) and the direct $|1\rangle \rightarrow |0\rangle$ transition (b) at $\eta = 0.3$ (dashed lines) and $\eta = 0.3 + 0.01$ (solid lines). (c) Intensity (integrated spectrum) of Stokes and direct peak. (d) Fisher information of the Stokes and direct peak. Parameters: $\omega_{\rm L} = 1.1\omega_c$

processes that we introduce in this work has important implications for the spectroscopic characterization of cavity QED systems via the analysis of excitation-emission spectra, contributing substantially to the amount of information that these measurements provide about the internal parameters of the system. In other words, we can significantly increase the precision by which internal parameters are estimated by making sure that not only the spectral features related to *direct* transitions between eigenstates are correctly fitted by our cavity-QED model, but that also Raman scattering peaks are.

In order to give a quantitative support to these claims, we analyze this question from the perspective of quantum parameter estimation [17–24]. As a relevant example, we consider the problem in which, assuming that our system is described by the quantum Rabi model Hamiltonian described in the main text, we have the task of estimating the light-matter coupling parameter η from the measurement of a emission spectrum.

Any given measurement strategy is described by a positive operator-valued mesaurement (POVM) Λ , i.e., a set of operators $\{\hat{\Lambda}_{\mu}\}$, where $\mu \in \{1, 2, ..., M\}$ labels different possible measurement outcomes, and $\sum_{\mu} \hat{\Lambda}_{\mu} = \mathbb{1}$. For a given value of our unknown parameter η , the probability for each of the different measurement outcomes follows a distribution $P(\mu|\eta) = \text{Tr}[\rho_{\eta}\hat{\Lambda}_{\mu}]$, where ρ_{η} is the η -dependent density matrix of the system at the time of the measurement. The uncertainty in the estimation of η from this measurement, $\Delta^2 \eta$, is bounded by means of the Cramér-Rao bound by the Fisher information of the probability distribution, $\Delta^2 \eta \ge 1/F$, which is defined as

$$F = \mathbf{E}\left[\left(\frac{d\log P(\mu|\eta)}{d\eta}\right)^2\right].$$
(S14)

In Ref. [25], an expression for the Fisher information associated to measurements of emission spectra was derived. This was done by describing the measurement of the emission spectrum $S(\omega)$ as a collection of independent measurements over a discretized set of N frequency points $\vec{\omega} = [\omega_1, \omega_2, \dots, \omega_N]$, which can be arbitrarily close and that would span over the measured frequency range, each point corresponding to the value measured through a different frequency filter. Assuming that, for each frequency point ω_i the probability distribution of the measurement of the spectrum is a Poissonian distribution with mean value $S(\omega_i)$, the Fisher information thus reads

$$F = \sum_{i=1}^{N} \frac{1}{S(\omega_i, \eta)} \left[\frac{\partial S(\omega_i, \eta)}{\partial \eta} \right]^2,$$
(S15)

By applying the Cramér-Rao bound, this quantity allows us to evaluate the metrological potential of spectrum measurements for the estimation of an unknown η . Also, since F is given by a sum of independent terms, we can isolate the contribution to the Fisher information from different regions of the spectrum. Here, we will do so, separating the contributions to the Fisher information from the spectrum measured in two different frequency ranges; one centered around the frequency of a direct transition from the eigenstate $|1\rangle$ to the ground state $|0\rangle$, and one centered around the frequency of the Raman peak corresponding to the Stokes process $|0\rangle \rightarrow |1\rangle$, whose frequency we label as $\omega_{\rm S}^{(10)}$ in the main text. The spectral peaks measured in both frequency ranges are displayed in Fig. S1(a-b), where we used the same parameters for the calculation of the spectrum of Fig. 1(b) in the main text ($\eta = 0.3$. $\omega_{\rm L} = 1.1\omega_c$). A small change in η imprints a change in these spectral forms (see difference between solid and dashed lines in Fig. S1(a-b). Our calculation shows that the total intensity of the Raman peak (spectrum integrated over the considered frequency range) is 20% the intensity of the direct peak. In contrast, its Fisher information is 44% the contribution of the direct peak.

This example illustrates how the Raman peaks emerging in the ultra strong coupling regime of cavity QED contain a significant amount of information about the internal parameters of the system. Therefore, their correct description and understanding is of paramount importance for a correct characterization of cavity QED setups via spectroscopic measurements.

The example also highlights a major difference between Raman photons spontaneously scattered by cavity QED systems and Raman photons scattered via the coupling to phononic degrees of freedom. In the case without phonons, the process is completely described by a quantum Rabi Hamiltonian, and all the features in the spectrum can be fitted by such a model (e.g., the central frequency is completely determined by the transition energies between different eigenstates of the quantum Rabi Hamiltonian). However, in the case of Raman scattering due to coupling to phonons, the frequency of i.e., a scattered Stokes photon, ω_S , would be shifted from the laser frequency ω_L by an amount given by the frequency ω_m of the phonon mode:

$$\omega_{\rm S} = \omega_{\rm L} - \omega_m,\tag{S16}$$

which is a parameter independent of the cavity QED system. This means that it would not be possible to fit such a peak with a quantum Rabi model alone. This important different between both cases allows us to determine the origin of the observed Raman peaks, and unambiguously identify the situations in which these originate from the purely cavity QED effect that we report in this work.

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