The nature of the Lamb shift in weakly-anharmonic atoms: from normal mode splitting to quantum fluctuations

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When a two level system (TLS) is coupled to an electromagnetic resonator, its transition frequency changes in response to the quantum vacuum fluctuations of the electromagnetic field, a phenomenon known as the Lamb shift. Remarkably, by replacing the TLS by a harmonic oscillator, normal mode splitting leads to a similar shift, despite its completely classical origin. In a weakly-anharmonic system, lying in between the harmonic oscillator and a TLS, the origins of such shifts can be unclear. An example of such a system is the transmon qubit in a typical circuit quantum electrodynamics setting. Although often referred to as a Lamb shift, it cannot originate purely from vacuum fluctuations since in the limit of zero anharmonicity, the system becomes classical. Here, we treat normal-mode splitting separately from quantum effects in the Hamiltonian of a weakly-anharmonic system, providing a framework for understanding the extent to which the frequency shift can be attributed to quantum fluctuations.

Quantum theory predicts that vacuum is never at rest. On average, the electromagnetic field of vacuum has no amplitude, quantum vacuum fluctuations, however, impose a fundamental uncertainty in its value. This is captured in its most elementary form as the ground-state energy of a harmonic oscillator $\hbar \omega_r/2$. When an atom couples off-resonantly to a single electromagnetic mode, which is equivalent to a harmonic oscillator, the quantum vacuum fluctuations of the mode cause shifts in the transition frequencies between states of the atom. This effect was first measured by Lamb and Rutherford [1]. If the atom can be modeled as a two level system (TLS), this interaction is described in the rotating wave approximation (RWA) by the Jaynes-Cummings Hamiltonian [2]. The so-called Lamb shift is then given by $-g^2/\Delta$ where $g$ is the coupling strength and $\Delta = \omega_r - \omega_a$ is the frequency detuning between the mode ($\omega_r$) and atom ($\omega_a$). When two harmonic oscillators are coupled to each other classically, a quantitatively similar effect occurs from normal-mode splitting, where in the dispersive regime, each oscillator acquires a frequency shift due to the presence of the other oscillator. In the RWA, this shift has the same magnitude $-g^2/\Delta$ despite its different, and classical origin. A system which lies between the harmonic oscillator and the two level system, a weakly-anharmonic atom characterized by a small anharmonicity $\lambda$, will also be shifted in frequency by the same magnitude. It has also been attributed to vacuum fluctuations and referred to as the Lamb shift [3]. In the limit $\lambda \rightarrow 0$, however, the shift has to converge to the classical effect of normal-mode splitting. To what extent then is the Lamb shift of a weakly-anharmonic atom induced by quantum vacuum fluctuations?

Here, we find that for a weakly-anharmonic atom coupled dispersively to a harmonic oscillator, two distinct shifts occur; one is a quantum effect due to vacuum fluctuations, another is classical in its nature and identical to normal-mode splitting. To illustrate the described physics, this work focuses on the transmon qubit [4] coupled to a LC-circuit in a typical circuit quantum electrodynamics situation. We follow the approach of transforming the Hamiltonian to its normal-mode basis [5]. We then treat anharmonicity as a perturbation, distinguishing the classical effect of normal-mode splitting from the quantum effects resulting from the anharmonicity. By performing these calculations analytically, we gain insight into the origin of different frequency shifts, and reach accurate approximations of their magnitude, extending the expressions previously derived through perturbation theory to regimes of large detuning [4].

We use a transmon qubit as an example of a physical implementation of a weakly-anharmonic artificial atom. We start by examining the energy shifts that arise from quantum fluctuations of current in the isolated atom. Applying the same concepts to the atom coupled to a resonator, we separate energy shifts in the form of classical normal mode splitting from those with a purely quantum origin. The latter being induced by vacuum fluctuations of the current in the resonator, we refer to this shift as the Lamb shift. The analysis is first limited to the rotating wave approximation regime, where $g \ll |\Delta| \ll \Sigma \ (\Sigma = \omega_r + \omega_a)$, before being extended to all dispersive regimes $g \ll |\Delta|, \Sigma$.

We define a weakly-anharmonic atom as a harmonic oscillator with a small quartic potential following the Hamiltonian

$$\hat{H}/\hbar = \omega_a (\hat{a}^\dagger \hat{a} + 1/2) - \frac{\lambda}{12} (\hat{a} - \hat{a}^\dagger)^4,$$

where $\hat{a}$ is the annihilation operator for excitations in the atom, $\omega_a$ the atomic frequency and $\lambda$ the anharmonicity. We can verify that this Hamiltonian has a weakly-anharmonic level structure by applying pertur-
bination theory. In the limit $\lambda \ll \omega_a$, the correction to the eigen-energies of $\hat{H}_\text{HO}$ due to the anharmonicity is to first order equal to $-\left(\lambda/12\right)\langle n \rangle \langle \hat{a} - \hat{a}^\dagger \rangle^4\langle n \rangle$, with $|n\rangle$ a number state. We can expand $\langle \hat{a} - \hat{a}^\dagger \rangle^4$ and only consider terms that preserve the number of excitations $n$, since only they will give a non-zero contribution to the first-order correction

$$
\hat{H}_\text{anh}/\hbar \simeq -\frac{\lambda}{2} \left( \langle \hat{a}^\dagger \hat{a} \rangle^2 + \hat{a}^\dagger \hat{a} + \frac{1}{2} \right).
$$

This leads to the energy levels

$$
E_n/\hbar \simeq (\omega_a - \lambda) \left( n + \frac{1}{2} \right) - \lambda \left( \frac{n^2}{2} - \frac{n}{2} - \frac{1}{4} \right).
$$

If we write the transition frequencies of the atom $E_n - E_{n-1} = \omega_a - n\lambda$, the slightly anharmonic level structure becomes apparent and is illustrated in the energy diagram of Fig. 1(a).

We will use the transmon qubit as an example of weakly-anharmonic atom [4]. In addition to being described by the simple electrical circuit depicted in Fig 1(a), this system is highly relevant in many experimental endeavors [6], ranging from fundamental experiments in quantum optics [7–11], to quantum simulations [12] or the development of a quantum processor [13–15]. The transmon qubit is an artificial atom constructed from an LC oscillator where the linear inductance is replaced by the non-linear inductance $L_J(I)$ of a Josephson junction (JJ). We will use the notation $L_J = L_J(0)$.

The limit of weak anharmonicity for a transmon qubit translates to the zero-point fluctuations in current being much smaller than the junctions critical current $I_c$. In this limit, the current $I$ traversing the JJ when only a few excitations populate the circuit is much smaller than the critical current of the JJ and $L_J(I) \simeq L_J(1 + I^2/2I_c^2)$. Intuitively, the expectation value of the current squared $\langle I^2 \rangle$, on which the inductance depends, will increase with the number of excitations in the circuit. So with increasing number of excitations $n$ in the circuit, the effective inductance of the circuit increases and the energy of each photon number state $E_n$ will tend to decrease with respect to the harmonic case. For a rigorous quantum description of the system, the flux $\phi(t) = \int_0^t V(t')dt'$, where $V$ is the voltage across the JJ, is a more practical variable to use than current [16], and using the conjugate variables of flux and charge the Hamiltonian of Eq. (1) can be shown to describe the transmon [4]. The anhar-
monicity is given by the charging energy $\hbar \lambda = e^2/2C$, the atomic frequency by $\omega_a = 1/\sqrt{LJC}$ and the flux relates to the annihilation operator through $\phi = \phi_{\text{zpf}} (\hat{a} - \hat{a}^\dagger)$, where the zero-point fluctuations in flux are given by $\phi_{\text{zpf}} = \sqrt{\hbar L_J/C/2}$. Note that for a linear inductance $L$ the flux $\phi$ is proportional to the current $I$ traversing the inductor $\phi = LI$. We can recover the intuition gained by describing the system with currents by plotting the eigen-states of the harmonic oscillator in Fig. 1(a). In the normalized flux basis $\hat{\phi} = \phi/\phi_{\text{zpf}}$, we see that the fluctuations of the eigen-states increases with the excitation number, and hence the expectation value of the fourth-power of the flux $\langle \hat{\phi}^4 \rangle \propto \langle \hat{H}_{\text{anh}} \rangle$ will increase. The energy of each eigen-state will then decrease, slightly deviating from a harmonic level structure.

We now study the effect of coupling a harmonic oscillator to the atom. When an $LC$ oscillator is connected capacitively to a transmon (see Fig. 1(b)), the following Hamiltonian can be derived through circuit quantization [4, 16]

$$\hat{H}/\hbar = (\omega_a + \lambda) \hat{a}^\dagger \hat{a} - \frac{\lambda}{12} (\hat{a} - \hat{a}^\dagger)^4 + \omega_r \hat{b}^\dagger \hat{b} + g (\hat{a} + \hat{a}^\dagger) (\hat{b} + \hat{b}^\dagger).$$  \hspace{1cm} (4)

Here $\hat{b}$ is the annihilation operator of photons in the resonator, $\omega_r$ is its frequency and $g$ the coupling strength. Compared to the Hamiltonian of Eq. (1), we replaced the frequency $\omega_a$ scaling the atomic number operator with $\omega_a + \lambda$. Doing so will ensure that $\omega_a$ corresponds to the frequency of the first atomic transition, independent of the anharmonicity $\lambda$, as proven by Eq. (3). We also omitted the ground-state energies $\hbar \omega_r/2$ and $\hbar (\omega_a + \lambda)/2$ in this Hamiltonian; even though vacuum fluctuations are at the origin of these omitted terms, their presence plays no role in calculating the transition frequencies of the system and the Lamb shift.

To describe the dispersive regime $g \ll |\Delta|$ of this interaction, we want to distinguish the energy shifts due to normal-mode splitting, that would be present even with $\lambda = 0$, from the shifts originating from quantum fluctuations. To do so, we first move to the normal-mode basis, as described in Refs. [17, 18] and Supplementary Sec. 1 [19]. By introducing the normal-mode frequencies $\tilde{\omega}_a = \omega_a - \delta_{\text{NM}}$ and new operators $\tilde{\alpha}, \tilde{\beta}$ which eliminate the coupling term in Eq. (4) whilst preserving the usual commutation relations

$$\hat{H}/\hbar = (\tilde{\omega}_a + \lambda) \tilde{\alpha}^\dagger \tilde{\alpha} + \tilde{\omega}_r \tilde{\beta}^\dagger \tilde{\beta} - \frac{1}{12} \left( \chi_a^{1/4} (\tilde{\alpha} - \tilde{\alpha}^\dagger) + \chi_r^{1/4} (\tilde{\beta} - \tilde{\beta}^\dagger) \right)^4. \hspace{1cm} (5)$$

These new operators $\tilde{\alpha}, \tilde{\beta}$ have a linear relation to $\hat{a}, \hat{b}$, which determines the value of $\chi_a$ and $\chi_r$ (see Supplementary Sec. [19]). Expanding the anharmonicity leads to

$$\hat{H}_{\text{anh}}/\hbar = -\frac{\chi_a}{2} \left( \hat{\alpha}^\dagger \hat{\alpha}^2 + \hat{\alpha} \hat{\alpha}^\dagger + \frac{1}{2} \right) - \frac{\chi_r}{2} \left( \hat{\beta}^\dagger \hat{\beta}^2 + \hat{\beta} \hat{\beta}^\dagger + \frac{1}{2} \right) - 2\chi_{ar} \left( \hat{\alpha}^\dagger \hat{\alpha} + \frac{1}{2} \right) \left( \hat{\beta}^\dagger \hat{\beta} + \frac{1}{2} \right), \hspace{1cm} (6)$$

if we neglect terms which do not preserve excitation number and are therefore irrelevant to first order in $\lambda$. This approximation is valid for $\lambda \ll |\Delta|, |3\omega_a - \omega_r|, |\omega_a - 3\omega_r|$, which notably excludes the straddling regime that was thoroughly studied in Ref. [4]. The anharmonicity (or self-Kerr) of the normal-mode splitted atom and resonator $\chi_a$ and $\chi_r$ is related to the AC Stark shift (or cross-Kerr) $2\chi_{ar}$ through

$$\chi_{ar} = \sqrt{\chi_a \chi_r}. \hspace{1cm} (7)$$

The AC Stark shift is the change in frequency one mode acquires as a function of the number of excitations in the other. The presence of this term, as well as an anharmonicity of the resonator, can be understood from the mechanism of normal-mode splitting. When the transmon and $LC$ oscillator are dispersively coupled, the normal-mode corresponding to excitations in the $LC$ oscillator will be composed of currents oscillating through its inductor but also partly through the JJ. We can then decompose the current $I$ traversing the JJ into two contributions, the current corresponding to atomic excitations $I_a$ and resonator excitations $I_r$. In Eq. (5), this effect appears in the terms of flux as $\phi = \phi_a + \phi_r \propto \chi_a^{1/4} (\hat{\alpha} - \hat{\alpha}^\dagger) + \chi_r^{1/4} (\hat{\beta} - \hat{\beta}^\dagger)$. Consequently, the value of the JJ inductance is now not only dependent on the number of excitations in the atom but also in the resonator: the JJ inductance increases with the number of excitations in the resonator.

The dependence of the JJ inductance on the current of the resonator mode has two consequences. The resonator will acquire a small anharmonicity, following the same formula as Eq. (3) with $\lambda = \chi_r$. Additionally, the frequency of the atom is now dependent on the number of excitations in the resonator. Even when the resonator is in its ground state, there is a shift due to the vacuum current fluctuations of the resonator. This can be verified by the presence of $1/2$ in the cross-Kerr term of Eq. (6) which arise as a consequence of the commutation relations $[\hat{\alpha}, \hat{\alpha}^\dagger] = [\hat{\beta}, \hat{\beta}^\dagger] = 1$, mathematically at the origin of vacuum fluctuations. Since this shift is the only one which arises from the vacuum fluctuations of the coupled oscillator, we will call it the Lamb shift.

To summarize, compared to an isolated harmonic oscillator the energy levels of the atom are shifted by three distinct effects: (1) the classical normal-mode splitting
\(\lambda g\phi\delta\omega a\) its own anharmonicity \(\chi a\) which arises from the quantum fluctuations of its own eigen-states, and (3) the Lamb or AC Stark shift arising from the quantum fluctuations of the resonator it is coupled to. These different effects are depicted in Fig. 1(b). In Fig. 2(a,b), we show how these shifts manifest in a typical experimental setting where the detuning between the atom and resonator is varied, without explicitly showing contribution (2). Beyond the point of resonance, both modes are slightly shifted with respect to their un-coupled frequencies, and our theory allows us to distinguish the different effects which contribute to this shift.

In the rotating wave approximation (RWA) regime \(g \ll |\Delta| \ll \Sigma\), the following approximations hold

\[
\begin{align*}
\bar{\omega}_a &\approx \omega_a - g^2 \bar{\Delta} - \lambda g^2 \bar{\Delta}^2, \\
\bar{\omega}_r &\approx \omega_r + g^2 \bar{\Delta} + \lambda g^2 \bar{\Delta}^2, \\
\chi a &\approx \lambda \left(1 - 2g^2 \bar{\Delta}^2\right), \\
\chi r &\approx \mathcal{O}(g^4), \\
\chi ar &\approx \lambda g^2 \bar{\Delta}^2,
\end{align*}
\]

valid to leading order in \(g\) and \(\lambda\). The expression for the AC Stark shift was also derived by Koch \textit{et al.} [4] from perturbation theory, given in the form \(\lambda g^2 / \Delta (\Delta - \lambda)\). Applying perturbation theory to the Hamiltonian of Eq. (4) is however less accurate and provides less physical insight. Namely it fails to predict the correct shift beyond the RWA regime and it does not make the distinction between the physical origin of the different shifts.

From Eqs. (8), we find that the total shift acquired when the resonator is in its ground-state \(\delta\omega a = \lambda - \delta NM - \chi a - \chi ar\), is equal to \(-g^2 / \Delta\). This shift is equal to that of a harmonic oscillator coupled to another harmonic oscillator (here, the case \(\lambda = 0\)) as well as that of a TLS coupled to a harmonic oscillator. The fact that the total shift has the same magnitude in these three different systems can easily lead to a confusion as to its origin. In particular since the shift of a TLS is a purely quantum effect, that of two coupled harmonic oscillators is a purely classical effect, and the weakly-anharmonic system lies somewhere in between. This confusion can now be addressed: for a weakly-anharmonic system, there is a contribution from normal-mode splitting and a contribution from vacuum fluctuations which can both be quantified, and the former is much larger than the latter for a weakly-anharmonic atom like the transmon. Note that here, the Lamb shift refers to only the quantum-fluctuations-induced part of the total dispersive shift of the atom, in contrast to terminology used previously [5, 17]. This also explains why earlier work [3] found the Stark shift per photon to be smaller than what was referred to at the time as the Lamb shift: vacuum fluctuations was not the only measured effect, normal-mode splitting also greatly contributed to the measured shift. The proportion to which the total shift is due to vacuum fluctuations, as a function of anharmonicity, is
FIG. 3. Lamb shift beyond the RWA regime fixing $\lambda/\omega_a = 0.01$ and $g/\omega_a = 0.02$. A numerical calculation (half the shift from adding a photon in the oscillator), shown as the full red line, is compared to our analytical expression valid beyond the RWA $\chi_{ar} = 4 \lambda g^2 \omega_a^2/\Delta^2 \Sigma^2$, shown as the dashed blue line. The expression for the Lamb shift valid in the RWA is shown in dashed dashed green, which is also the expression derived by Koch et al. [4]. The resonances at which our theory is invalid are denoted by red shaded bars, at $\omega_r = \omega_a$ and $\omega_r = 3 \omega_a$ when the neglected term proportional to $(\beta - \beta')(\hat{a} - \hat{a}')^3$ comes into play.

shown in Fig. 2(c).

Beyond the RWA to regimes of large detuning $g \ll |\Delta| \sim \Sigma$ the approximate expressions of the different shifts are given by

\[
\begin{align*}
\bar{\omega}_a &\simeq \omega_a - g^2 \frac{2 \omega_r}{\Delta \Sigma} - 4 \lambda g^2 \frac{\omega_r \omega_a}{\Delta^2 \Sigma^2}, \\
\bar{\omega}_r &\simeq \omega_r + g^2 \frac{2 \omega_a}{\Delta \Sigma} + 4 \lambda g^2 \frac{\omega_a^2}{\Delta^2 \Sigma^2}, \\
\chi_a &\simeq \lambda \left(1 - 4 g^2 \frac{\omega_a}{\omega_r} \left(\frac{\omega_a^2 + \omega_r^2}{\omega_a^2 \Delta^2 \Sigma^2}\right)^2 \right), \\
\chi_r & = \mathcal{O}(g^4), \\
\chi_{ar} & \simeq 4 \lambda g^2 \frac{\omega_a^2}{\Delta^2 \Sigma^2}.
\end{align*}
\]

An important difference with the RWA case is that the AC Stark shift $2 \chi_{ar}$ scales with $\omega_r^2$, decreasing with the frequency of a coupled resonator as shown in Fig. 3. This notably explains why the transmon qubit is insensitive to low frequency charge fluctuations as compared to the highly anharmonic Cooper pair box. It also explains why the transmon qubit is not adapted to measuring individual quanta of far-off-resonant systems such as low frequency mechanical oscillators [20]. Contrary to the AC Stark shift in the RWA, this expression cannot be derived by applying perturbation theory to Eq. (4). The different shifts which arise from this method and perturbation theory are compared to two coupled harmonic oscillators and the two level system case in Supplementary Table S1 and Fig. S2 [19].

In conclusion, we have presented a method to separate the classical effects of normal-mode splitting from the consequences of quantum fluctuations in the Hamiltonian of a weakly-anharmonic atom coupled to a harmonic oscillator. Through our theory, we reveal the physical origin of the different energy shifts arising in such a system. The main result is that only a small fraction of the total frequency shift acquired by a weakly-anharmonic atom can be attributed to quantum vacuum fluctuations, the dominant part of the shift is due to classical normal-mode splitting. In addition to addressing this fundamental question, we expect that the expressions derived in Eqs. (8) and (9), as well as our approach to studying this Hamiltonian will become practical tools for experimental efforts in circuit QED.

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The nature of the Lamb shift in weakly anharmonic atoms: from normal mode splitting to quantum fluctuations

Supplemental Material

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S1. TRANSFORMATION TO THE NORMAL-MODE BASIS

The Hamiltonian

$$\hat{H}/\hbar = (\omega_a + \lambda) \hat{a}^{\dagger} \hat{a} + \omega_r \hat{b}^{\dagger} \hat{b} + g(\hat{a} + \hat{a}^{\dagger})(\hat{b} + \hat{b}^{\dagger})$$

(S1)

describes two harmonic oscillators with a linear interaction between them. It can be compactly written as

$$\hat{H}/\hbar = v^T H v,$$

$$v^T = [\hat{a}, \hat{b}, \hat{a}^{\dagger}, \hat{b}^{\dagger}],$$

$$H = \frac{1}{2} \begin{bmatrix} 0 & g & (\omega_a + \lambda) & g \\ g & 0 & g & \omega_r \\ (\omega_a + \lambda) & g & 0 & g \\ g & \omega_r & g & 0 \end{bmatrix},$$

(S2)

omitting constant contributions. Using this notation, the canonical commutation relations read

$$[v, v^T] = vv^T - (vv^T)^T = J = \begin{bmatrix} 0 & I_2 \\ -I_2 & 0 \end{bmatrix},$$

(S3)

where $I_2$ is the $2 \times 2$ identity matrix. The objective of this section is to rewrite (S1) as the Hamiltonian of two independent harmonic oscillators, or normal-modes

$$\hat{H}/\hbar = (\bar{\omega}_a + \lambda) \hat{\alpha}^{\dagger} \hat{\alpha} + \bar{\omega}_r \hat{\beta}^{\dagger} \hat{\beta},$$

(S4)

which we write in compact notation as

$$\hat{H}/\hbar = \eta^T \Lambda \eta,$$

$$\eta^T = [\hat{\alpha}, \hat{\beta}, \hat{\alpha}^{\dagger}, \hat{\beta}^{\dagger}]$$

$$\Lambda = \frac{1}{2} \begin{bmatrix} 0 & 0 & (\bar{\omega}_a + \lambda) & 0 \\ 0 & 0 & 0 & \bar{\omega}_r \\ (\bar{\omega}_a + \lambda) & 0 & 0 & 0 \\ 0 & \bar{\omega}_r & 0 & 0 \end{bmatrix},$$

(S5)

To do so, we need to find a matrix which maps $v$ to a new set of annihilation and creation operators of the normal-modes $\eta$ which should also satisfy the commutation relations (S3). The matrix $\Lambda J$ is diagonal

$$\Lambda J = \frac{1}{2} \begin{bmatrix} -(\bar{\omega}_a + \lambda) & 0 & 0 & 0 \\ 0 & -\bar{\omega}_r & 0 & 0 \\ 0 & 0 & (\bar{\omega}_a + \lambda) & 0 \\ 0 & 0 & 0 & \bar{\omega}_r \end{bmatrix},$$

(S6)

and we define it as the diagonal form of the matrix $HJ$. In other words, we can determine the value of $\bar{\omega}_a$ and $\bar{\omega}_r$ by diagonalizing $HJ$. As we will now demonstrate, defining $\Lambda$ in this way will lead to operators with the correct commutation relations. We define the matrix of eigen-vectors that diagonalizes $HJ$ as $F = [w_0, w_1, w_2, w_3]$, such that

$$HJ = F \Lambda J F^{-1}$$

(S7)
The matrix $F$ can be normalized in such a way that it satisfies an important condition, it can be made symplectic

$$F^T J F = F J F^T = J .$$  \hfill (S8)

If the eigenvectors are normalized such that $w_i^T w_i = 1$, the operation that leads to symplecticity is

$$w'_0 = \pm w_0 / \sqrt{|w_0^T J w_2|} ,$$

$$w'_1 = \pm w_1 / \sqrt{|w_1^T J w_3|} ,$$

$$w'_2 = \pm w_2 / \sqrt{|w_0^T J w_2|} ,$$

$$w'_3 = \pm w_3 / \sqrt{|w_1^T J w_3|} ,$$ \hfill (S9)

where the $+$ or $-$ sign is chosen such that if we redefine $F = [w'_0, w'_1, w'_2, w'_3]$ it is of the form

$$F = \begin{bmatrix} A & B \\ B & A \end{bmatrix} ,$$ \hfill (S10)

and such that $F = I_4$ in the limit $g = 0$. With $F$ a symplectic matrix, we can define $\eta$ as

$$\eta = F^T v$$ \hfill (S11)

and (Proposition 1) $\eta$ will respect the commutation relations (S3) whilst ensuring that (Proposition 2) the two Hamiltonians (S2) and (S5) are equivalent. Proof of these proposition is provided at the end of this section. With the relation (S15), we can invert (S11) to obtain

$$v = -J F J \eta .$$ \hfill (S12)

An exact expression for the normal-mode frequencies is given by

$$\bar{\omega}_{ar} = \frac{1}{\sqrt{2}} \left( (\omega_a + \lambda)^2 + \omega_r^2 \pm \sqrt{((\omega_a + \lambda)^2 - \omega_r^2)^2 + 16g^2(\omega_a + \lambda)\omega_r} \right) .$$ \hfill (S13)

Using the software Mathematica, we diagonalize $H J$ and perform the normalizations of Eqs. (S9); by Taylor expanding the resulting expressions for small values of $g$, we obtain

$$\bar{\omega}_a = \omega_a - \frac{2g^2 \omega_r}{\Sigma' \Delta'} ,$$

$$\bar{\omega}_r = \omega_r + \frac{2g^2 \omega_a}{\Sigma' \Delta'} ,$$

$$\dot{\alpha} \approx \left( 1 - g^2 \frac{2(\omega_a + \lambda)\omega_r}{\Delta'^2 \Sigma'^2} \right) \dot{\alpha} + g^2 \frac{\omega_r}{(\omega_a + \lambda) \Sigma' \Delta'} \dot{\beta} + g^2 \frac{\omega_r}{\Sigma' \Delta'} \dot{\beta} \hfill \frac{1}{\Sigma' \Delta'} \dot{\alpha}^{\dagger} - \frac{g}{\Sigma'} \dot{\beta}^{\dagger} ,$$ \hfill (S14)

$$\dot{\beta} \approx -g \frac{\Delta}{\Sigma'} \dot{\alpha} + \left( 1 - g^2 \frac{2(\omega_a + \lambda)\omega_r}{\Delta'^2 \Sigma'^2} \right) \dot{\beta} + g^2 \frac{\omega_a + \lambda}{\omega_r} \frac{1}{\Sigma' \Delta'} \dot{\alpha}^{\dagger} + g^2 \frac{(\omega_a + \lambda)}{\omega_r} \frac{1}{\Sigma' \Delta'} \dot{\beta}^{\dagger} ,$$

valid to second order in $g$ where $\Delta' = \Delta - \lambda$ and $\Sigma' = \Sigma + \lambda$. The above relations lead to the expressions for $\chi_a$ and $\chi_r$ given in the main text.
Proposition 1 – Proof Multiplying Eq. (S8) with $J$, we find

$$-FJF^T J = -JFJF^T = -J^2 = I_4 , \tag{S15}$$

where $I_4$ is the $4 \times 4$ identity matrix.

$$[\eta, \eta^T] = \eta\eta^T - (\eta\eta^T)^T \tag{S11}$$

$$\equiv F^T (vv^T) F - F^T (vv^T)^T F \tag{S16}$$

$$= F^T [v, v^T] F$$

$$\equiv F^T JF \tag{S3}$$

$$\equiv J , \tag{S8}$$

This proof illustrates how essential it is that $F$ be symplectic (Eq. (S8)) to obtain the desired commutators.

Proposition 2 – Proof The relation (S15) allows us to introduce the matrix $F$ into Eq. (S2)

$$\frac{\dot{H}}{\hbar} = v^T Hv \tag{S15}$$

$$\equiv -v^T H J F J F^T v \tag{S17}$$

$$= -v^T F A J F^{-1} F J F^T v$$

$$= I_4$$

$$\equiv -v^T F A J F^T v$$

$$= -I_4$$

$$= (F^T v)^T A (F^T v) ,$$

proving that $\frac{\dot{H}}{\hbar} = \eta^T A \eta$. 
FIG. S1. Domain of validity of the approximations made in this work. At two different detunings, the exactly calculated Lamb shift (half the shift from adding a photon in the oscillator), shown as a full orange line, is compared to the approximate expression for $\chi_{ar}$. When not varied, we fix $\lambda/\omega_a = 0.01$ and $g/\omega_a = 0.02$. For increasing $g$, we find that a second order approximation leads to correct predictions up to the ultra-strong coupling regime $g/\omega_a \simeq 0.1$. The range of valid anharmonicity is more limited, particularly for negative detuning $\omega_r < \omega_a$. For $\lambda/\omega_a > 0.02$, a significant deviation is observed and going to next order in $\lambda$ would be required. This involves second-order perturbation theory in the quartic term of Eq. (4) as well as going to another order in the expansion of the Josephson energy for the case of the transmon, leading to a term in $(x(\hat{\alpha} - \hat{\alpha}^\dagger) + y(\hat{\beta} - \hat{\beta}^\dagger))^6$. 
Model | $\chi_{ar}$ | $\delta\omega_a$
---|---|---
**With RWA**
two-level system | $\frac{g^2}{\Delta}$ | $-\frac{g^2}{\Delta}$
perturbation theory | $\lambda \frac{g^2}{\Delta^2}$ | $-\frac{g^2}{\Delta}$
our theory | $\lambda \frac{g^2}{\Delta^2}$ | $-\frac{g^2}{\Delta}$
harmonic oscillator | 0 | $-\frac{g^2}{\Delta}$

**Without RWA**
two-level system | $\frac{g^2}{\Delta} + \frac{g^2}{\Sigma}$ | $-\frac{g^2}{\Delta} + \frac{g^2}{\Sigma}$
perturbation theory | $\lambda \left( \frac{g^2}{\Delta^2} + \frac{g^2}{\Sigma^2} \right)$ | $-\frac{g^2}{\Delta} - 2\frac{g^2}{\Sigma} - \lambda + \frac{g^2}{\Sigma}$
our theory | $4\lambda g^2 \frac{\omega_r^2}{\Delta^2\Sigma^2}$ | $-\frac{g^2}{\Delta} - \frac{g^2}{\Sigma} + \lambda g^2 \frac{4\omega_r^2}{\omega_a \Delta^2\Sigma^2}$
harmonic oscillator | 0 | $-\frac{g^2}{\Delta} - \frac{g^2}{\Sigma}$

**TABLE S1.** Summary of the shifts of different models. Here, $\chi_{ar}$ refers to the Lamb shift, or half the AC Stark shift, and $\delta\omega_a$ to the total frequency shift of the ground to first excited state transition of the atom. The latter is the sum of the normal-mode shift $\delta_{NM}$, the Kerr shift $\chi_a$ and the Lamb shift $\chi_{ar}$, excluding the Kerr shift of the uncoupled atom $\lambda$: $\delta\omega_a = \lambda - \delta_{NM} - \chi_a - \chi_{ar}$. Perturbation theory refers to the application of quantum perturbation theory directly to the Hamiltonian of Eq. (3). The resonances at which our theory is invalid are denoted by red shaded bars, at $\omega_r = \omega_a$ and $\omega_r = 3\omega_a$ when the neglected term proportional to $(\hat{\beta} - \hat{\beta}^\dagger)(\hat{\alpha} - \hat{\alpha}^\dagger)^3$ comes into play.
FIG. S2. Illustration of the different approximations presented in Table S1. (a) As in Fig. 2(b) of the main text, we plot the total frequency shift $\delta \omega_a$, as a function of detuning $\Delta = \omega_r - \omega_a$. (b) As in Fig. 3, we plot the Lamb shift $\chi_{ar}$ as a function of detuning. The numerical calculation corresponds to half the shift from adding a photon in the coupled oscillator.