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Rabi-vibronic resonance with large number of vibrational quanta

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We study theoretically the Rabi oscillations of a resonantly driven two-level system linearly coupled to a harmonic oscillator (vibrational mode) with frequency, \( \omega_0 \). We show that for weak coupling, \( \omega_p \ll \omega_0 \), where \( \omega_p \) is the polaronic shift, Rabi oscillations are strongly modified in the vicinity of the Rabi-vibronic resonance \( \Omega_R = \omega_0 \), where \( \Omega_R \) is the Rabi frequency. The width of the resonance is \( (\Omega_R - \omega_0) \sim \omega_p^{2/3} \omega_0^{1/3} \gg \omega_p \). Within this domain of \( \Omega_R \) the actual frequency of the Rabi oscillations exhibits a bistable behavior as a function of \( \Omega_R \). Importantly, within the resonant domain, the oscillator is highly excited, which allows to treat it classically.

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I. INTRODUCTION

Coupling to environment tends to damp the Rabi oscillations\(^1\) of a resonantly driven two-level system. Usually, the environment is viewed as medium with continuous spectrum of modes. Less common is the situation when environment possesses a single or several well-defined frequencies. For concreteness we will consider the situation depicted in Fig. 1 when the lower level of the two-level system is coupled to an oscillator (a mass, \( M \), and a spring), which represents a single vibrational mode. Obviously, coupling to the oscillator has a strong effect on the Rabi oscillations in the regime of the vacuum Rabi splitting\(^2\) when the oscillator frequency, \( \omega_0 \), is close to the transition frequency, \( \omega_{12} \). It is less obvious what effect the coupling to the oscillator will have on the Rabi oscillations when \( \omega_0 \) is much smaller than \( \omega_{12} \) and is comparable to the Rabi frequency, \( \Omega_R \). One can argue on physical grounds that the effect of coupling on the Rabi oscillations will be strong in the vicinity of the condition, \( \omega_0 \approx \Omega_R \), which we dub Rabi-vibronic resonance. Indeed, consider the Hamiltonian

\[
H = \mu \hat{X} \hat{n}_1, \quad (1)
\]

describing the linear coupling. Here \( \hat{X} = \frac{1}{\sqrt{2M\omega_0}}(b + b^\dagger) \) is the operator of the oscillator displacement, \( b^\dagger \) is a creation operator of the vibrational quantum, \( \hat{n}_1 \) is the occupation of the level \( E_1 \), and \( \mu = (2M\omega_0^3)^{1/2} \lambda \), where \( \lambda \) is a dimensionless coupling constant. In definition of \( \hat{X} \), \( \mu \), and thereafter we set \( \hbar = 1 \). In the course of the Rabi oscillations the average \( \hat{n}_1 \) changes with time as

\[
n_1(t) = \frac{1}{2}(1 + \cos \Omega_R t). \quad (2)
\]

Then at \( \Omega_R \approx \omega_0 \), the second term in Eq. (2) gives rise to a resonant driving force acting on the oscillator.

FIG. 1: A schematic illustration of the system under consideration. Two-level system is driven by near-resonant light, \( \omega_{12} \approx (E_2 - E_1) \). The level \( E_1 \) is linearly coupled to a classical oscillator with frequency \( \omega_0 \). The Rabi oscillations are strongly modified when \( \omega_0 \) is close to \( \Omega_R \), where \( \Omega_R \) is the Rabi frequency.

In turn, the strongly driven oscillator provides a resonant feedback\(^3,4\) on the two-level system. Thus, as \( \Omega_R \), which is proportional to the ac field driving the two-level system, increases, we expect the Rabi oscillations to be strongly modified near the resonant condition.

Among possible experimental realizations of the situation Fig. 1 is a suspended carbon nanotube in inhomogeneous electric field, which creates a confinement for an exciton\(^5-8\). Localized exciton can be viewed as a two-level system. Bending modes having discrete frequencies due to finite nanotube length, can be viewed as oscillators with very low friction. While a typical transition frequency in such a system is\(^9,10\) \( \omega_{12} \sim 10^{15}\)Hz, the oscillator frequency\(^7\) is much smaller \( \omega_0 \sim 10^9\)Hz. Resonant condition can be achieved by adjusting the illumination intensity.

Another area in which the situation Fig. 1 is relevant, is the cavity QED\(^11\), where a two-level system is realized in
the form of a superconducting qubit, while the oscillator is a LC-circuit. Majority of experimental and theoretical studies in this field are focused on the strong coupling in the domain $\omega_0 \approx \omega_{12}$. However, in experiments Refs. [12,13] an ac driven superconducting qubit was coupled to a “slow” LC-oscillator tuned to $\Omega_R$. It was observed that the nonresonant spectrum of the oscillator exhibits a Lorentzian peak\textsuperscript{14} as a function of $\Omega_R - \omega_0$. In theoretical papers\textsuperscript{15–17} initiated by the experiment Ref. 12 collective motion of the oscillator coupled to a qubit was studied within the density matrix formalism, and both subsystems were treated quantum-mechanically. In view of complexity of this description, final results were obtained numerically for particular values of a coupling parameter, $\lambda$. A notable finding of Refs. [15–17] is that, in the vicinity of the condition $\omega_0 \approx \Omega_R$, collective Rabi-vibronic motion becomes bistable.

There are still several basic questions to be answered, among which:

(i) how the frequency, $s$, of the collective oscillations depends on $\lambda$?

(ii) what is the width of the resonance, i.e., the domain $\delta_0 = \Omega_R - \omega_0$ of the Rabi frequencies where Rabi oscillations are modified due to coupling?

(iii) how the decay of the Rabi oscillations depends on the oscillator friction?

The above questions are studied in the present paper. Our main finding is that the width of the Rabi-vibronic resonance is small for weak coupling, namely,

$$\delta_0 = \lambda^{4/3} \omega_0 = \omega_p^{2/3} \omega_0^{1/3} \ll \omega_0, \quad (3)$$

where $\omega_p = \lambda^2 \omega_0$ is the polaronic shift. Eq. (3) suggests that, while $\delta_0$ is much smaller than $\omega_0$, it is much bigger than $\omega_p$. Most importantly, Eq. (3) guarantees that, in the resonant domain $(\Omega_R - \omega_0) \sim \delta_0$, the oscillator is highly excited and can be treated as classical. This allows the analytical description of the resonance. In this regard, the situation we consider, two-level system coupled to a classical oscillator, is similar to the Rabi resonance considered in Refs. [18,19], where two-level system was driven by two classical fields: one with frequency close to $\omega_{12}$ and one with frequency close to $\Omega_R$.

We will see that in the domain $(\Omega_R - \omega_0) \sim \delta_0$, the frequency $s$ of collective oscillations differs from $\Omega_R$ also by $\sim \delta_0$. Bistable behavior of the dependence $s(\Omega_R)$ emerges naturally within our approach; the frequency jump rate between two stable regimes are also $\sim \delta_0$. In addition, in the present paper we study how the Rabi-vibronic resonance depends on detuning, $\Delta$, of the driving frequency from $\omega_{12}$, on intrinsic anharmonicity of the oscillator, and how the modified Rabi oscillations decay with time due to relaxation of the two-level system and due to friction in the oscillator.

**II. BASIC EQUATIONS**

We first assume that the displacement, $X(t)$, is a classical variable and will later check this assumption. The equation of motion for $X(t)$ reads

$$\ddot{X} + \gamma \dot{X} + \omega_0^2 X = \frac{\mu}{2M} (1 - w), \quad (4)$$

where $w = 1 - 2n_1$ is the population inversion, and $\gamma$ is the friction in the oscillator. The evolution of $w$ with time is described by the system of optical Bloch equations. We write them for variables $w(t)$, $u(t)$, and $v(t)$, where $u(t)$ and $v(t)$ are the real and imaginary parts of the nondiagonal elements of the density matrix, respectively\textsuperscript{20},

$$\dot{w}(t) = -\Omega_R v - \Gamma (1 + w), \quad (5)$$

$$\dot{u}(t) = -\left(\Delta - \mu X(t)\right) v - \frac{\Gamma}{2} u, \quad (6)$$

$$\dot{v}(t) = \left(\Delta - \mu X(t)\right) u + \Omega_R w - \frac{\Gamma}{2} v, \quad (7)$$

where $\Gamma$ is the relaxation rate of the excited state. Note that, while the oscillator is driven by $w(t)$, it exercises a feedback on the two-level system via $u(t)$ and $v(t)$.

We require that the level $E_1$ at $t = 0$ is occupied while the level $E_2$ is empty, i.e., $w(0) = -1$. We also assume that the dipole moment and dipole current are initially zero, leading to $v(0) = 0$ and $u(0) = 0$, respectively. From Eq. (5), we see that the initial conditions for $v$ and $w$ require that $\dot{w}(0) = 0$.

**III. MODIFIED RABI OSCILLATIONS**

**A. Oscillation frequency**

The system Eqs. (5)-(7) can be reduced to two coupled equations by excluding $v(t)$ and expressing $u(t)$ in terms of $w(t)$. Then one gets

$$\ddot{w} + \frac{3}{2} \Gamma \dot{w} + \left(\Omega_R^2 + \frac{\Gamma^2}{2}\right) w + \frac{\Gamma^2}{2} = -\Omega_R \left(\Delta - \mu X(t)\right) u(t), \quad (8)$$

$$u(t) = -\int_0^t \frac{dt'}{\Omega_R} e^{-\Gamma(t'-t)/2} \left(\mu X(t') - \Delta\right) \left[\dot{w}(t') + \Gamma (1 + w)\right]. \quad (9)$$
We start from the simplest case, $\Gamma \to 0$, $\Delta \to 0$, $\gamma \to 0$, and search for a solution of the system Eqs. (4), (8), and (9), in the form $w(t) = -\cos st$. Substituting this form into Eq. (4) we find the displacement

$$X(t) = \frac{\mu \cos st}{2M(\omega_0^2 - s^2)} = X_0 \cos st.$$  \hspace{1cm} (10)

Static displacement, $\mu/2M\omega_0^2$, can be neglected compared to the oscillating part. Substituting $X(t)$ into Eq. (9), we find $u(t)$

$$u(t) = \frac{\mu^2 (1 - \cos 2st)}{8M\Omega_R(\omega_0^2 - s^2)}. \hspace{1cm} (11)$$

Substituting Eqs. (10), (11) into the right-hand side of Eq. (8), and equating the terms $\propto \cos st$ in both sides, we find a closed equation for $s$

$$\Omega_R^2 - s^2 = \frac{\omega_0^2\omega_4}{8(\omega_0^2 - s^2)^2}. \hspace{1cm} (12)$$

This expression is valid only if the correction on the right-hand side is much smaller than $(\omega_0 - \Omega_R)$. Equating the correction to $(\omega_0 - \Omega_R)$, we find that the width of the resonance, $(\omega_0 - \Omega_R) \sim \delta_0$, is given by Eq. (3).

Recall now our basic assumption that the oscillator is classical. We are now in position to verify this assumption. In the resonant domain the amplitude, $X(t)$, can be estimated from Eq. (10) as $X \sim \mu/\omega_0\delta_0$. Then for the ratio of the energy of oscillations to the vibrational quantum, $\omega_0$, we get the following estimate

$$MX_0^2 \omega_0^2 \sim \left(\frac{\omega_0}{\omega_p}\right)^{1/3} = \lambda^{2/3} \gg 1. \hspace{1cm} (14)$$

Thus, for weak coupling, the classical treatment of the oscillator is justified.

![Figure 2](image-url)  

**FIG. 2:** Red line: Dimensionless frequency, $z$, defined by Eq. (17), of Eq. (19) versus the dimensionless deviation, $x$, from the $\Omega_R = \omega_0$. Unstable solution is shown with dashed line. Blue line, corresponds to the absence of coupling to the oscillator.

**B. Vicinity of the resonance**

To incorporate finite detuning, $\Delta$, into Eq. (12) it is convenient to rewrite Eq. (8) keeping all $\Delta$-dependent terms in the right-hand side

$$\ddot{w} + \Omega_R^2 w + \mu^2 X(t) \int_0^t dt' X(t') \dot{w}(t') = \Delta (\mu X(t) - \Delta) [w(t) + 1] + \Delta \mu \int_0^t dt' X(t') \dot{w}(t'). \hspace{1cm} (15)$$

The term $\propto \Delta^2$ in the right-hand side leads to a standard modification of the Rabi frequency to $(\Omega_R^2 + \Delta^2)^{1/2}$. The last term is proportional to $\sin^2 st$, and does not contain the first harmonics. The term $\propto \cos st$ comes from the combination $\Delta \mu X(t)$ in the right-hand side. Emergence of this term, which is odd in detuning, is the result of the coupling of the vibronic mode only to the level $E_1$. This term results in the following modification of Eq. (12)

$$\Omega_R^2 + \Delta^2 - s^2 = \frac{\omega_0^2\omega_4}{8(\omega_0^2 - s^2)^2} - \frac{\omega_p\omega_0^2\Delta}{\omega_0^2 - s^2}. \hspace{1cm} (16)$$

Near the resonance $(\Omega_R - \omega_0) \ll \omega_0$ this equation can be simplified. Upon introducing dimensionless variables,

$$z = \frac{s - \omega_0}{\omega_p^{1/3} \omega_0^{1/3}}, \hspace{1cm} (17)$$

$$x = x' + 8\Delta^2, \hspace{0.5cm} x' = \frac{\Omega_R - \omega_0}{\omega_p^{1/3} \omega_0^{1/3}}. \hspace{1cm} (18)$$
The last factor in Eq. (21) is the amplitude of a zero-point motion of the oscillator. As the dimensionless deviation, \( x \), from resonance increases, the upper branch approaches \( z = x \). For this branch the frequency of the Rabi oscillations is close to \( \Omega_R \) and the amplitude of oscillator is small. For the lower branch \( z \) is small, i.e., the frequency of the oscillations approaches \( \omega_0 \) with increasing \( x \). For this branch the oscillator is highly excited.

Figs. 3 and 4 illustrate the effect of detuning on the frequency of oscillations, \( s \). Note that there is a qualitative difference between Fig. 2 for zero detuning, and Figs. 3 and 4 for positive and negative detunings, respectively. For zero detuning, the domain of bistability exists only when \( \Omega_R > \omega_0 \), whereas for finite detuning, bistable regions emerge both to the left and the right from the resonance. As one changes the dimensionless deviation, \( x' \), from the resonance, from negative to positive, for \( \Delta' = 0 \), bistability corresponds to \( x' > 3 \cdot 2^{-8/3} \). For finite positive detuning, \( \Delta' > 0 \), the first domain of bistability occurs at \( x' < 0 \), then disappears, and reemerges at positive \( x' \) greater than \( 3 \cdot 2^{-8/3} \). Conversely, finite negative detuning simply broadens the domain of bistability as compared to \( \Delta = 0 \). Bistable region starts for \( x' < 0 \). Peculiar dependence of \( s \) on the deviation from resonance is also reflected in the amplitude of the oscillator. This effect is discussed in the Sect. V.

C. Effect of intrinsic anharmonicity of the oscillator

Suppose that in addition to the harmonic part of the oscillator energy, \( M \omega_0^2 X^2/2 \), a weak intrinsic anhar-
monicity, κX^4/4, is present. Then Eq. (10) will assume the form
\[ 3 \kappa X_a^3 + (\omega_0^2 - s^2) X_a = \frac{\mu}{2M}. \]  
(22)

The second relation, \( s^2 = \Omega_R^2 - \mu^2 X_a^2 / 8 \), between the amplitude, \( X_a \), and the frequency, \( s \), which follows from Eq. (8) remains unchanged. It is now more convenient to express \( s \) from this relation and substitute it into Eq. (22). This yields a cubic equation for \( X_a \).

\[ 2\omega_0 (\omega_0 - \Omega_R) X_a + \frac{6\kappa + \mu^2}{8} X_a^3 = \frac{\mu}{2M}. \]  
(23)

If we now set \( \kappa = 0 \), then Eq. (23) will have multiple real solutions for \( X_a \) in the domain \( (\Omega_R - \omega_0) < -3 \cdot 2^{-8/3} \delta_0 \), i.e., the same as determined from Eq. (19) with \( \Delta' = 0 \). We see from Eq. (23) that, depending on the sign of \( \kappa \), intrinsic anharmonicity can either shift the threshold of bistability to the left (for positive \( \kappa \)) or to the right (for negative \( \kappa \)). Anharmonicity will also affect the magnitude of the jump of the frequency, \( s \), of the oscillations. This magnitude will get modified from 2^{-8/3} \delta_0 to 2^{-8/3} \delta_0 (1 + 6 \kappa / \mu^2)^{-2/3}, i.e., the jump will become smaller for \( \kappa < 0 \).

IV. DECAY OF THE OSCILLATIONS

Up to now we disregarded both mechanisms of dissipation: finite relaxation, \( \Gamma \), and the friction in the oscillator, \( \gamma \). Rabi oscillations will decay with the rate \( \Upsilon \), which is determined by \( \Gamma \), in the regime \( \Gamma \gg \gamma \), or by friction in the regime \( \Gamma \ll \gamma \). We will consider both cases separately. We emphasize that, as the oscillations decay, so does the coupling between the oscillator and two-level system. Thus the decay will be accompanied by the change of frequency back to \( \Omega_R \). We cannot capture this evolution of frequency with time analytically. To find the decay rate, \( \Upsilon \), only, we will adopt the approach based on the energy conservation.

A. Friction-dominated regime, \( \gamma \gg \Gamma \)

Upon neglecting \( \Gamma \) in Eq. (8) and setting \( \Delta = 0 \) we have
\[ \dot{\omega} + \Omega_R^2 \omega = -\mu^2 X(t) \int_0^t dt' X(t') \dot{\omega}(t'). \]  
(24)

Multiplying both sides by \( \dot{\omega} \) and integrating from 0 to \( t \), we arrive to the following conservation law
\[ \frac{\dot{\omega}^2}{2} + \frac{\Omega_R^2}{2} (\omega^2 - 1) = -\frac{\mu^2}{2} \left( \int_0^t dt' X(t') \dot{\omega}(t') \right)^2. \]  
(25)

The right-hand side describes the energy exchange between the two-level system and the oscillator.

As a next step we multiply the equation of motion of the oscillator Eq. (4), by \( X \) and integrate from 0 to \( t \). Then we arrive at the second conservation law
\[ \frac{\dot{X}^2}{2} + \frac{\omega_0^2}{2} X^2 + \gamma \int_0^t dt' \dot{X}^2(t') = -\frac{\mu}{2M} \int_0^t dt' \dot{w}(t') \dot{X}(t'). \]  
(26)

At long time, \( t \gg \Upsilon^{-1} \) we have, \( \dot{\omega}, \dot{w} \to 0 \) and also \( X, \rightarrow 0 \). Then the left-hand side of Eq. (25) for \( w \) turns to \(-\Omega_R^2 / 2 \). Combining Eqs. (25) and (26), we arrive to the relation
\[ \Omega_R = 2M \gamma \int_0^\infty dt' \dot{X}^2(t'). \]  
(27)

This relation is convenient to find the decay rate, \( \Upsilon \), because it contains only \( \dot{X}^2 \), which is insensitive to the change of frequency in the course of decay. Substituting \( \dot{X}^2 \propto \exp[-2\Upsilon t] \), we find the following relation between \( \Upsilon \) and the amplitude of the oscillations at time \( t \leq 1/\Upsilon \)
\[ \Upsilon = \gamma (M \dot{X}_0^2 \Omega_R). \]  
(28)

In fact, Eq. (28) does not prove that decay is exponential. In the next subsection we will see that it becomes exponential only for long times, \( \Upsilon t \gg 1 \). Note that the second factor in the right-hand side can be rewritten as \((M \dot{X}_0^2 \Omega_R^2)/\Omega_R \). The oscillator is classical when this ratio is big. Thus we conclude that \( \Upsilon \gg \gamma \), i.e., the Rabi oscillations in the region of resonance decay faster than undriven oscillator. As a next step, we distinguish two cases of weak and strong friction. In the first case, to find \( X \) that should be substituted into Eq. (28) one can use Eq. (10) obtained without friction. Then one gets
\[ \Upsilon = \frac{\gamma \omega_p \Omega_R}{(\omega_0 - \omega)^3}, \]  
(29)

where \( s \) is determined from the cubic equation Eq (13). In the region of resonance, the difference \( \omega_0 - s \) is \( \sim \delta_0 \), which yields
\[ \Upsilon \sim \gamma \left( \frac{\omega_0}{\omega_p} \right)^{1/3} = \frac{\gamma}{\lambda^{2/3}}. \]  
(30)

Weak friction requires that \( |\omega_0 - s| \sim \delta_0 \gg \Upsilon \), i.e., \( \gamma \ll \omega_p \).

In the region of strong friction the difference \( \omega_0 - s \) should be replaced by \( \Upsilon \). Then Eq. (29) contains \( \Upsilon \) in both sides. Upon solving this equation, we get
\[ \Upsilon \sim \left( \frac{\gamma}{\omega_p} \right)^{1/3} \delta_0, \]  
(31)
for $\gamma \gg \omega_p$. Equations (30), (31) match when $\gamma \sim \omega_p$. The validity of this expression is limited from above by the condition that the oscillator is classical. As we replace $\omega_0 - s$ by $\Upsilon$, the estimate for $X$ is $X \sim \mu/(M\omega_0\Upsilon)$. Then the kinetic energy can be estimated as

$$M\omega_0^2 \left[ \frac{\mu}{M\omega_0\Upsilon} \right]^2 \sim \frac{\omega_p^{1/3}}{\omega_0^{4/3}} \frac{\gamma^{2/3}}{\Upsilon^{2/3}}.$$  

(32)

The condition that it is bigger than $\omega_0$ limits $\gamma$ in Eq. (31) to $\gamma \ll (\omega_0\omega_p)^{1/2} = \lambda_0\omega_0$, and correspondingly limits $\Upsilon$ to $\Upsilon \ll \delta_0/(\omega_0/\omega_p)^{1/6} = \delta_0/\lambda^{1/3}$.

From Eqs. (30), (31) we see that, upon increasing friction, the decay rate, $\Upsilon$, first grows linearly with $\gamma$, and then sublinearly, as $\gamma^{1/3}$. At the boundary of applicability of the classical description we have $\Upsilon = \gamma$. For even bigger $\gamma$ classical treatment of the oscillator is not justified, but we expect that the oscillator will eventually decouple from the two-level system, and Rabi oscillations will proceed as they do in the absence of the oscillator.

It is convenient to reformulate the above results in terms of number, $m = \omega_0/\Upsilon$, of the oscillation cycles after which the collective motion effective stops. From Eqs. (30), (31) we have

$$m = \begin{cases} \lambda^{2/3} \left( \frac{\omega_p}{\gamma} \right), & \gamma < \lambda^2\omega_0 \\ \frac{1}{\lambda^{2/3}} \left( \frac{\omega_p}{\gamma} \right)^{1/3}, & \lambda_0\omega_0 > \gamma > \lambda^2\omega_0 \end{cases}$$  

(33)

In the crossover between weak and strong friction regimes we have $m \sim \lambda^{-4/3}$. For $\gamma > \lambda_0\omega_0$ the assumption of classical motion of the oscillator is violated. The boundary value of $m$ at $\gamma \sim \lambda_0\omega_0$ is still large, $m \sim \lambda^{-1}$.

### B. The form of the decay

For more quantitative analysis of the decay of oscillations, it is convenient to rewrite the energy conservation law Eq. (25) in terms of the displacement, $X(t)$. Expressing $w(t)$ from Eq. (4) and substituting it into Eq. (25) we get

$$\left[ \frac{d}{dt}(\ddot{X} + \gamma\dot{X} + \omega_0^2 X) \right]^2 + \Omega_R^2(\ddot{X} + \gamma\dot{X} + \omega_0^2 X)^2 \approx \frac{\mu^2\Omega_R^2}{4M^2} - \mu^2 \left( \int_0^t \left[ X\ddot{X} + \omega_0^2 X\dot{X} + \gamma X\dot{X} \right] \right)^2.$$  

(34)

The first two terms in the integrand can be presented in the form

$$X\ddot{X} + \omega_0^2 X\dot{X} = \frac{d}{dt} \left[ X\ddot{X} - \frac{\dot{X}^2}{2} + \frac{\omega_0^2 X^2}{2} \right],$$  

(35)

so that the integral from this terms is equal to $\frac{1}{2}(\omega_0^2 - s^2)X^2(t)$. At the same time, the integral from the third term can be rewritten as $\frac{1}{2}\gamma s^2 \int_0^t dt' X^2(t')$, and estimated at $\gamma s^2 X_0^2/\Upsilon$. One can check using Eqs. (30), (31) that both in the strong-friction and weak-friction regimes the integral from the last term is bigger than the contribution from the first two terms. Neglecting this contribution, we can cast Eq. (34) in the form of an integral equation for the slow decaying amplitude, $\tilde{X}$, of the oscillations.

Since $\gamma \ll |\omega_0 - s|$ in both regimes, the non-oscillating part of the left-hand side of Eq. (34) can presented as

$$\frac{1}{2}(\omega_0^2 - s^2)(s + \Omega_R^2)\tilde{X}^2 \approx \frac{\mu^2\omega_0^2\tilde{X}^2}{4M^2\omega_0^2},$$  

(36)

where we used the definition Eq. (10) of the initial amplitude, $X_0$. Upon substituting Eq. (36) into Eq. (34) and introducing a dimensionless function $F(t) = \tilde{X}(t)/X_0$, we arrive at the integral equation

$$F^2(t) = 1 - \Upsilon \left[ \int_0^t dt' F(t') \right]^2,$$  

(37)

where $\Upsilon = \gamma M\omega_0^{1/2}$ is introduced according to Eq. (29). It is easy to check that this equation has a simple solution

$$F(t) = \frac{1}{\cosh \Upsilon t}.$$  

(38)

We see that the decay of amplitude $\tilde{X}(t)$ becomes exponential in the limit $\Upsilon t \gg 1$, as mentioned in the previous subsection.

### C. Initial stage of the oscillations

After the ac driving field is switched on, the population inversion starts to oscillate with frequency $\Omega_R$. After some number, $m'$, of the oscillation cycles the frequency crosses over to $s$. The question of interest is how $m'$ depends on the coupling strength, $\lambda$. We will estimate $m'$ using the fact that at initial stage the system Eqs. (4), (24) can be solved perturbatively in small parameter $\omega_p/\omega_0$. To find the perturbative solution, we substitute the “bare” Rabi oscillations $w^{(0)} = -\cos(\Omega_R t)$ into Eq. (4) and find $X(t)$ with initial conditions $X(0) = 0$, $\dot{X}(0) = 0$. The obtained $X(t)$ together with $w^{(0)}(t)$ is then substituted into the right-hand side of Eq. (24). Solving this second-order differential equation with a given right-hand side, we find that the amplitude of oscillations becomes $1 - w^{(1)}(t)$, i.e., it acquires a time-dependent correction with $w^{(1)}(t)$ given by the following
expression

\[
w^{(1)}(t) = \frac{2\omega_p^2\omega_0^2}{(\gamma^2 + 4\delta^2)^2}\left[\frac{\gamma t}{2} - \frac{4\gamma\delta}{(\gamma^2 + \delta^2)}e^{-\gamma t/2}\sin \delta t - \frac{\gamma^2 - 4\delta^2}{\gamma^2 + 4\delta^2}\left(1 - e^{-\gamma t/2}\cos \delta t\right)\right]^2,
\]

(39)

where \(\delta = \omega_0 - \Omega_R\). We can now estimate \(m'\) as \(\Omega_R t_c \approx \omega_0 t_c\), where \(t_c\) is the time after which \(w^{(1)}(t)\) becomes \(\sim 1\). Consider first the limit \(\delta t \rightarrow 0\). This yields

\[m' \sim \frac{\gamma}{\omega_0\lambda^2}.
\]

(40)

Condition that \(m' \gg 1\) should be consistent with the condition that the oscillator is classical. The latter condition reads: \(\omega_p\omega_0 \gg \gamma^2\), which is equivalent to \(\gamma \ll \lambda\omega_0\). In the domain when both conditions are met we have \(1 \ll m' \ll \frac{1}{\lambda}\).

Consider now the limit \(\delta t \ll 1\). For \(\delta t \ll 1\), Eq. (39) yields

\[w^{(1)}(t) \sim (\omega_p\omega_0 t^2)^2.\]

This leads to the estimate \(t_c \sim (\omega_p\omega_0)^{-1/2}\) and \(m' \sim \lambda^{-1}\). Small-\(t\) expansion of Eq. (39) is valid if the product \(\delta t_c\) is small. With \(t_c\) found above, this product can be rewritten in the form

\[\delta t_c \sim \left(\frac{\delta^2}{\omega_p\omega_0}\right)^{1/2}.
\]

(41)

On the other hand, the oscillator can be treated as classical when the ratio in the right-hand side of Eq. (41) is small. Thus, taking the limit \(\delta t \ll 1\) in Eq. (39) is justified, and the frequency of the Rabi oscillations crosses over from \(\Omega_R\) to \(s\) after \(m' \sim \lambda^{-1}\) cycles. Correspondingly, after \(\sim \lambda^{-1}\) cycles, the oscillator will "forget" about initial phase, imposed by the initial conditions, and will execute a forced harmonic motion with frequency, \(s\).

In conclusion of this subsection we note that for the entire scenario of the collective oscillations to be consistent the time during which the collective motion is established must be shorter than the time during which these oscillations decay. The corresponding condition is \(m' < m\). It follows from Eqs. (33), (40) that this condition is satisfied in the the entire interval \(\gamma < \lambda\omega_0\), namely, \(m\) is always bigger than \(\lambda^{-1}\), while \(m'\) is always smaller than \(\lambda^{-1}\).

D. Relaxation-dominated regime, \(\Gamma \gg \gamma\)

At finite relaxation rate of the two-level system Eq. (26) assumes the form

\[
\frac{\dot{w}^2}{2} + \frac{1}{2}\left(\Omega_R^2 + \Gamma^2\right)(w^2 - 1) + \frac{\Gamma^2}{2}(w + 1) + \frac{3}{2}\Gamma\int_0^t dt' w^2 = -\mu^2\int_0^t dt' X(t')\dot{w}(t')\int_0^t dt'' e^{\Gamma/2}(t'' - t')/2 X(t'')|\dot{w}(t'') + \Gamma(1 + w)|.
\]

(42)

Without coupling to the oscillator the right-hand side is zero, and Eq. (42) describes the decay of the Rabi oscillations due to relaxation. Indeed, upon substituting \(\dot{w} = \Omega_R \sin(\Omega_R t')\exp(-\Gamma t')\) and taking the limit \(t \rightarrow \infty\), the last term in the left-hand side takes the value \(3\Gamma\Omega_R^2/8\Gamma\), which leads to \(\Upsilon = 3\Gamma/4\). Naturally, this value of \(\Upsilon\) follows directly from Eqs. (5), (7). Finite coupling to the oscillator would increase the decay rate only if at \(t \rightarrow \infty\) the integral in the right-hand side exceeds \(\Omega_R^2\). Contribution of the second term in the square brackets to the integral can be estimated upon noticing that the product \(X(t'')w(t'')\) is a slow function. Assuming that \(X\) and \(w\) both decay as \(\exp(-\Upsilon t'')\) and that \(\Upsilon \gg \Gamma\), the integral over \(t''\) reduces to \(\int_0^\infty dt'' \sin(2\Gamma t'')\exp(-2\Upsilon t'') = \Upsilon/\delta^3 \approx \Upsilon/\lambda^3\). Then one gets the estimate \(\omega_p^2\Upsilon/\left(\omega_0 - s\right)^2\) for this contribution. Since \(\omega_0 - s\) cannot be smaller than \(\Upsilon\), this contribution cannot exceed \(\omega_p^2\Upsilon^2/\lambda^6\), which is much smaller than \(\Omega_R^2\). The contribution from the first term in the square brackets also cannot exceed \(\Omega_R^2\). This becomes apparent upon performing integration by parts after which the contribution from the first term assumes the form

\[-\Gamma\mu^2\int_0^\infty dt' e^{-\Gamma t'}\left(\int_0^t dt'' e^{\Gamma t''/2} X(t'')\dot{w}(t'')\right)^2.
\]

(43)

If \(X\) and \(\dot{w}\) decay much faster than \(\exp(-\Gamma t'/4)\), the inner integral saturates at times \(t' \ll \Gamma\). Then the contribution Eq. (43) can be estimated as \(\Omega_R^2\omega_p^2/\left(\omega_0 - s\right)^2\), which is again much smaller than \(\Omega_R^2\). We thus conclude that,
expressed using Eq. (21), as the following
\[ N = 16N\lambda^{2/3}, \]
where \( N \) is the number of vibrational quanta, is plotted from Eq. (19) vs. dimensionless detuning and dimensionless deviation from the resonance, \( x' = (\Omega_R - \omega_0)/(\omega_0\omega_p^2)^{1/3} \). The thick black line of bifurcations separates the "inner" domain of parameters (blue domain where bistability is absent) and outer domain (yellow domain where bistability is present). In the outer domain the ratio of \( N \) to the vibrational quanta, \( N\lambda \), is plotted from the resonance and to the detuning, \( \Delta \). Since \( \Delta = x' \), and displacement, \( \omega_p \), satisfy the harmonic oscillator equations with frequencies \( \Omega_R \) and \( \omega_0 \), respectively. We studied the behavior of the frequency the Rabi oscillations near the resonance \( \Omega_R = \omega_0 \). The number of vibrational quanta, \( N \), is also sensitive to the deviation, \( \Omega_R - \omega_0 \), from the resonance and to the detuning, \( \Delta \). Since \( N = M\omega_0^2X^2/\omega_0 \) (we set \( \hbar = 1 \)) it can be expressed using Eq. (21), as the following
\[ N = \left( \frac{1}{\lambda^2} \right)^{1/3} \frac{1}{16z^2} = \left( \frac{\omega_0}{\omega_p} \right)^{1/3} \frac{1}{16z^2}, \]
where \( z \) is the solution of Eq. (19). The dependence of \( N \) on dimensionless deviation, \( x' \), and dimensionless detuning, \( \Delta' \), is plotted in Fig. 5. The values of \( N \) shown correspond only to stable regimes of oscillations. The line of bifurcation points separates the \( (x', \Delta') \) domains with and without bistability in Fig. 5. In the domain of bistability the higher and lower values of \( N \) coincide along the red line. Away from the red line the high-\( N \) and the low-\( N \) values differ very strongly. High-\( N \) values correspond to the regime of oscillations with frequency close to \( \omega_0 \), see Fig. 3, whereas low \( N \)-values correspond to the frequency of oscillations close to \( \Omega_R \).

At \( x' < 0.5 \) and to the right from the bifurcation line there is no bistability. The value of \( N \) is low in this domain around \( \Delta' = 0 \). As \( \Delta' \) increases, \( N \) grows for both signs of detuning, \( \Delta' \). However, for \( \Delta' > 0 \) (blue detuning) the growth of \( N \) is monotonical. At the same time, for \( \Delta' < 0 \) (red detuning) the bistability sets in at certain critical \( \Delta' \). Upon further increase of \(|\Delta'|\) the low-\( N \) value does not grow, while the high-\( N \) value grows rapidly.

It is instructive to compare the results shown in Fig. 5 to the results of Refs. [15,16]. The curves \( N(\Delta', \Omega_R) \) were obtained in Refs. [15,16] by numerical solution of the system of master equations for the density matrix describing both the two-level system and the oscillator. Firstly, there is a qualitative agreement in the shape of the boundary of bistability. In Refs. [15,16] only the low-\( N \)-values are plotted. The prime observation made in Refs. [15,16] was that there is a strong difference between these low-\( N \)-values for blue and red detunings, namely, for blue detuning \( N \) is much higher. Our analytical results in Fig. 5 agree qualitatively with this observation.

V. CONCLUDING REMARKS

Frequency, \( \Omega_R \), of the Rabi oscillations is proportional to the square root of the excitation power. This linearity has been demonstrated in many experiments. Even when Rabi oscillations are damped, the dependence \( s(\Omega_R) \) can be extracted from the position of maximum in the Fourier transform\(^{21}\) of the signal, \( w(t) \). We predict that, for a two-level system coupled to a vibrational mode, the position of maximum of the Fourier transform will deviate from the linear behavior near the resonance \( \Omega_R = \omega_0 \). Both to the left and to the right from the resonance the position of maxim corresponds to \( s < \Omega_R \). The relative width of the resonant region depends on the coupling, \( \lambda \), to the vibrational mode as \( \lambda^{2/3} \). We also predict that, in the vicinity of the resonance, the dependence \( s(\Omega_R) \) exhibits a hysteretic behavior with two stable values of \( s \) corresponding to two stable regimes of the Rabi oscillations.

The underlying physics of the Rabi-vibronic resonance is the following. Without coupling, population inversion, \( w \), and displacement, \( X \), satisfy the harmonic oscillator equations with frequencies \( \Omega_R \) and \( \omega_0 \), respectively. With coupling, two-level system acts as a driving force \( \propto w \) on

\[ x' = (\Omega_R - \omega_0)/(\omega_0\omega_p^2)^{1/3}, \]
\[ N = \left( \frac{1}{\lambda^2} \right)^{1/3} \frac{1}{16z^2} = \left( \frac{\omega_0}{\omega_p} \right)^{1/3} \frac{1}{16z^2}. \]
the oscillator, while the back-action of the oscillator on $w$ is peculiar. The structure of back-action force is $wX^2$, as can be seen from Eqs. (8), (9). This structure implies that back-action is of a parametrical type, i.e., $X^2$ adds to $\Omega_R^2$. Thus, at $\Omega_R \approx \omega_0$, it appears that $\Omega_R$ is modulated with frequency $\approx 2\Omega_R$. This, however, does not lead to a parametric instability. Instead, the oscillator motion gets synchronized with the Rabi oscillations. In this regard, there is certain analogy to the synchronization of the Rabi oscillations to a sequence of pulses applied to the detector with repetition period chosen to be $2\pi/\Omega_R$.

As it was pointed out in Introduction, the situation when a two-level system undergoing the Rabi oscillations is coupled to the oscillator is actively studied in connection to the circuit QED\(^1\). The most common situation in circuit QED is when the oscillator frequency, $\omega_0$, is tuned to the transition frequency, $\omega_{12}$, of the two-level system. Among physical effects predicted for this domain is that two or multiple qubits can get strongly coupled to each other via coupling to a common oscillator\(^\text{23,24}\). Rabi-vibronic resonance corresponds to the domain $\omega_0 \ll \omega_{12}$. Still the effects similar to those discussed in Refs. [23,24] (see also recent experiments Refs. [25,26]) will take place under the conditions of the Rabi-vibronic resonance. In particular, we anticipate that Rabi oscillations in two driven two-level systems with $\Omega_R = \omega_0$, coupled to the same oscillator will get synchronized.

As a final remark, classical treatment of the vibrational mode adopted in the present paper does not allow one to capture the quantum jumps\(^1\) between the stable regimes of collective motion of the two-level system coupled to the oscillator. We also did not consider the effect of thermal noise which leads to the activated switching\(^\text{27}\) between the steady regimes even within a classical description of the oscillator.

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