

Ultrafast Control and Rabi Oscillations of Polaritons

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We report the experimental observation and control of space and time-resolved light-matter Rabi oscillations in a microcavity. Our setup precision and the system coherence are so high that coherent control can be implemented with amplification or switching off of the oscillations and even erasing of the polariton density by optical pulses. The data are reproduced by a quantum optical model with excellent accuracy, providing new insights on the key components that rule the polariton dynamics.

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Rabi oscillations [1] are the embodiment of quantum interactions: when a mode a is excited and is coupled to a second mode b , the excitation is transferred from a to b and when the symmetric situation is established, the excitation comes back in a cyclical unitary flow. When this occurs at the single particle level between two-level systems, it provides the ground for qubits [2], which, if they can be further manipulated, opens the possibility to perform quantum information processing [3]. Such an oscillation is of probability amplitudes and therefore is a strongly quantum mechanical phenomenon, that involves maximally entangled states

$$|\Psi(t)\rangle = \alpha(t)|1_a, 0_b\rangle + \beta(t)|0_a, 1_b\rangle. \quad (1)$$

The same physics also holds, not at the quantum level, but with coherent states of the fields, a situation known in the literature as implementing an “optical atom” [4] or a “classical two-level system” [5]. The oscillation is then more properly qualified as “normal mode coupling” [6,7] as it is now between the fields themselves,

$$|\psi(t)\rangle = |\alpha(t)\rangle|\beta(t)\rangle, \quad (2)$$

rather than their probability amplitudes. The denomination of Rabi oscillations remains, however, popular also in this case [8,9]. While of limited value for hardcore implementation of quantum information processing, it is desirable for fundamental purposes and semiclassical applications to have access to such classical qubits, or “cebits” [10]. In particular, they can help to explore the origin and mechanism of nonlocality and parallelization in genuinely quantum systems [11], as well as providing classical counterparts useful for proof-of-principle demonstration, design, and optimization of the actual quantum version [12]. Such classical two-level systems have been pursued

for decades [13] and recently enjoyed a boost with the rise of nanomechanical optics [5,14]. There is another system which provides an ideal platform to implement both genuinely quantum [15] and classical versions [16] of the two-level system: polaritons [17]. A polariton is by essence a two-level system, arising from strong light-matter coupling between a cavity photon and a semiconductor exciton. In planar microcavities embedding inorganic quantum wells (QWs), which is the case of interest here, the system has enjoyed considerable attention for its quantum properties at the macroscopic level [18], such as Bose-Einstein condensation [19], superfluidity [20,21] and a wealth of quantum hydrodynamics features [22–25], culminating with the demonstration of possible devices [26,27] and pioneering logical operations [28]. While Rabi oscillations are at the heart of polariton physics, they are so fast in a typical microcavity—in the subpicosecond time range—that they are typically glossed over and the macroscopic physics of polaritons investigated in their coarse graining. Pioneering attempts to observe them showed the inherent difficulty and reported hardly two oscillations with 3 orders of magnitude loss of contrast each time [29], attributed to the inhomogeneous broadening of excitons by the theory [30], which could provide a qualitative agreement only. Later reports through pump-probe techniques [31–33], in particular, in conjunction with an applied magnetic field [34], increased their visibility but remained tightly constrained to their bare observation. Since polaritons are increasingly addressed at the single particle level [35,36], it becomes capital to harness their Rabi dynamics [37].

In this Letter, thanks to significant progress in both the quality of the structures (the sample description is given in the Supplemental Material [38]) and in the laboratory state of the art, we have been able to both neatly observe and control the microcavity polariton Rabi dynamics. This brings microcavities one step further as platforms to engineer various states of light-matter coupling. We can

span from Rabi oscillating configurations to eigenstate superpositions, and control them by optical pulses that can amplify or switch states, thereby achieving the same type of coherent control recently reported in mechanical systems [5], but fully optically and with over 9 orders of magnitude gain in speed. The data offer a perfect quantitative agreement with a fundamental model of light-matter coupling of two bosonic fields [47], that allows us to pin down the underlying dynamics and explain which factors play which role and to which extent, at the highest level of precision ever attained in a microcavity, thus making such systems even more suitable for engineering and applications.

A typical experimental observation is shown in Fig. 1(a): the cavity field oscillates after its excitation by a 130 fs long and 8 nm energy broad pulse impinging on both branches, as sketched in Fig. 1(b). The basic interpretation is straightforward: by exciting both branches, the system is prepared as a bare state and, not being an eigenstate, oscillates between its two components. Since polaritons are extended objects, the oscillation is between two fields, localized in a Gaussian of width a few tenths of a μm given by the exciting laser. One can access the complex wave function, i.e., measuring both its amplitude and phase, by holography, a technique of increasing use to image polariton fluids for which both of these components are of crucial importance [48,49]. We recourse to a variation known as off-axis digital holography [50], which provides high-quality results by separation of the diffracted images of an off-axis reference frame and the signal. We adapted it to support ultrafast and tunable multiple-pulse experiments, with an overall time resolution of 130 fs (cf. Supplemental Material [38]). As such, our measurement does not rely on nonlinear interactions, as in previous works [31–33], but on interferences only. The power was set to excite polaritons at a low enough density in order to maintain their

bosonic properties in the linear regime. We can thus observe the subpicosecond linear Rabi oscillations through the coherent fraction $|\psi_a(\mathbf{r}, t)|^2$ of the cavity field in both space \mathbf{r} and time t (see supplemental movie [41]).

Both the photon-field ψ_a dynamics of the experiment and the complementary exciton field ψ_b , not accessible experimentally, can be recovered by the usual polariton field equations [51] (cf. Supplemental Material [38]). As expected, the exciton field forms as the photon field vanishes before it is revived as the excitations flow back from excitons into photons again. Limiting this to cases with no momentum—although the wave function components have a spread in both real and reciprocal spaces—the system is linear and there is no dynamics imparted by the spatial degree of freedom. The dynamics can therefore be reduced to zero dimension between two single harmonic modes, and the oscillations are fully captured through the simpler order parameters $\langle a(t) \rangle = \langle \psi | a | \psi \rangle$, accessible experimentally, and $\langle b(t) \rangle = \langle \psi | b | \psi \rangle$. This is shown in Fig. 1(c) as points, now for the full duration of the experiment. Twelve oscillations are clearly resolved until $t = 10$ ps. Theoretically, the Hamiltonian is reduced to simply $H_0 = \hbar\omega_0(a^\dagger a + b^\dagger b) + \hbar g(a^\dagger b + ab^\dagger)$, with coupling strength g between the photonic mode a and the emitter annihilation operator b , both following Bose algebra and at energy ω_0 (resonant case), supplemented with $H_\Omega = \sum_{c=a,b} P_c(t) e^{i\omega_L t} e^{i\phi_c} c^\dagger + \text{H.c.}$ This is the most general case of coherent and resonant excitation, with coupling to both fields and allowing for a relative phase, which is necessary to reproduce the data. Although the excitation is an optical laser shone directly on the cavity, which is often described theoretically as a cavity-only coupling term [51–53], it is clear on physical grounds that such a general form may be required instead: since the exciton field would still be excited without the cavity, it is natural that part of the excitation is shared between the latter and the QWs. While it has little consequence for the single-pulse excitation, this will be crucial when dealing with coherent control by a second pulse. This prepares states of the type of Eq. (2) regardless of the magnitude of pumping, i.e., classical states that should not be confused with quantum superpositions of the type of Eq. (1) [54], which would be extremely difficult to realize and maintain even for small values of $|\alpha|^2$ and $|\beta|^2$. By integrating Schrödinger's equation $i\hbar\partial_t\psi = H\psi$, one easily finds the closed-form expression for $\alpha(t)$ and $\beta(t)$ under the dynamics of $H = H_0 + H_\Omega$ (see Supplemental Material [38]). For the case of an initial state $|\alpha_0\rangle|\beta_0\rangle$, $|\psi(t)\rangle$ reduces to $|\alpha_0 \cos(gt) - i\beta_0 \sin(gt)\rangle - i\alpha_0 \sin(gt) + \beta_0 \cos(gt)\rangle$. This describes two quantum oscillators, swinging like any other of their classical counterparts, and that mixes features of the bare states (which amplitudes oscillate), with those of the polaritons (with no oscillations of their amplitudes). From the observation of the oscillation alone, it is therefore difficult to capture the true dynamics at play. This is where a theoretical model is needed to shed light on the hidden

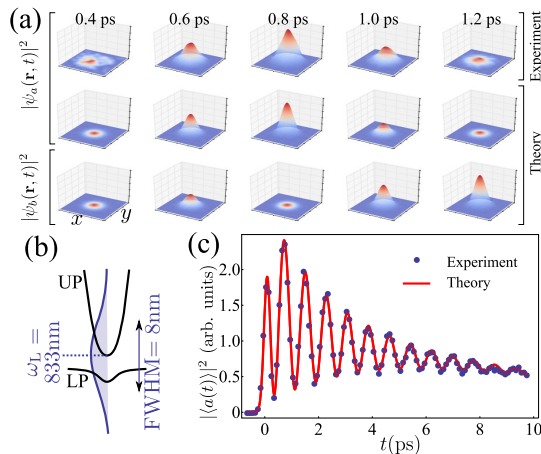


FIG. 1 (color online). (a) Oscillations observed experimentally in the cavity field and reproduced theoretically in both the cavity and exciton fields. (b) The lower (LP) and upper (UP) polaritons excited by a Gaussian pulse which overlap with the branches determine the effective state created in the system. (c) The dynamics can be reduced to that of $|\langle a(t) \rangle|^2$ alone and described quantitatively by the theory.

features [47]. As we are going to show, the contrast of the oscillation is not due to decoherence between the UP and LP, but to a combination of the short lifetime of the UP and of the effective state realized by the pulsed excitation.

While the core of the physics is contained in the wave function $|\psi\rangle$ of the coupled oscillators under the dynamics of H , we have to take into account dephasing and decay to describe any experiment with some degree of accuracy. These are mainly due to the bare state lifetimes (with decay rates $\gamma_{a,b}$ for the photon or exciton, respectively), which are also present in most light-matter coupled systems. In QW microcavities, additional sources of dephasing are present for the UP, which is notoriously less visible than its LP counterpart [55,56]. A contribution from the exciton reservoir has also been suggested in several works, even under coherent excitation [57,58]. However, no direct measurement of its contribution, nor its true nature (coherent or incoherent) has been clearly reported until now. We can address this issue by including an UP dephasing rate γ_U and an incoherent excitonic pumping rate P_b . Indeed, both terms are required to reproduce the data at the level of accuracy we report. Such terms turn the pure state wave function into a density matrix ρ ruled by a master equation. The theory is standard and is given in the Supplemental Material [38]. In this case, the complex amplitudes of the oscillators can also be derived in closed-form expressions. The experimental modulus square of the cavity amplitude can then be fitted by the model and other observables reconstructed from the theory. The fit provides an essentially perfect agreement with the data, as seen in the figures.

By shifting the laser energy to weight more on one branch than the other, as done for the series displayed in Fig. 2, different states can be prepared, which are all equally well accounted for by the theory for the same system parameters. Note also that both the dynamics of the pulse as well as the subsequent free oscillations are described within the same model. From the fit of the experimental $|\langle a(t) \rangle|^2$, we gain access to the entire dynamics of oscillations, also of the exciton field $|\langle b(t) \rangle|^2$, but even further, of the phases $\langle a(t) \rangle$ and $\langle b(t) \rangle$ and the total excitations $\langle (a^\dagger a)(t) \rangle$ and $\langle (b^\dagger b)(t) \rangle$ and, in fact, of the full state as a whole through the density matrix ρ . This allows us to reconstruct the full dynamics, as done in Fig. 2 for the joint exciton-photon oscillations of the experiment (case of 833 nm excitation), and see the effect of the various factors involved. For instance, the impact of the reservoir is seen in case I (in a dashed black line, from now on plotting only the envelope of the Rabi oscillations for clarity) where its effective pumping rate P_b has been set to zero. Its effect is small but is needed to reproduce the data quantitatively. The main detrimental actor is the UP dephasing rate γ_U , which, if set to zero, considerably opens the envelope of oscillations (case II, yellow dashed-dotted line). Interestingly, the incoherent reservoir extends the lifetime of the oscillations as shown in case II and even more so in case III (long dashed line) where a higher pumping rate than that of the experiment brings the oscillations well into the nanosecond time scale, as proposed in

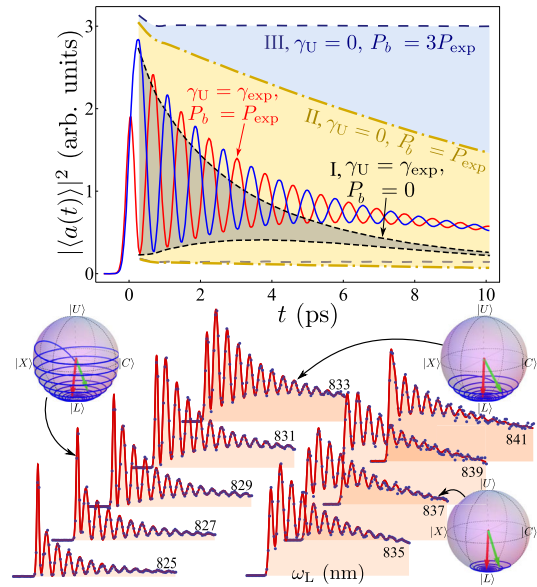


FIG. 2 (color online). Various states created in the system by varying the pulse energy and their evolution on the Bloch sphere, showing the systematic relaxation towards the LP. In the inset, the full exciton-photon dynamics are reconstructed theoretically for the case $\omega_L = 833$ nm (photon in red, exciton in blue) and variations (showing the envelope of the oscillations only) when removing the effect of the exciton reservoir (I, black dashed), removing the effect of polariton dephasing (II, dotted-dashed yellow) or, on the opposite, enhancing the effect of the reservoir (III, long-dashed blue). P_{exp} and γ_{exp} are the fitted values for the experiment.

Ref. [54], although, as already noted, this is for normal mode coupling oscillations that cannot be used to engineer a qubit.

The coherent amplitudes of any two-level system can be mapped on the Bloch sphere as $\langle a \rangle / \sqrt{|\langle a \rangle|^2 + |\langle b \rangle|^2} = \cos(\theta/2)$ and $\langle b \rangle / \sqrt{|\langle a \rangle|^2 + |\langle b \rangle|^2} = \sin(\theta/2) \exp(i\phi)$ with θ and ϕ the azimuthal and radial angles of polar coordinates, respectively. Such trajectories from our experiment are shown for three cases in Fig. 2, corresponding to predominant UP excitation, equal weight of the branches, and predominant LP excitation. It is clearly seen in the first case how the pulse swings the coupled oscillators towards the upper state and, in all cases, how the system quickly reaches the LP. This is the clearest observation to date of one of the most important assumptions of microcavity polariton physics: the UP is unstable and the system relaxes towards the lower branch, even though it retains strong coupling. In the model, this UP dephasing rate γ_U , could be either an escape rate (like a lifetime due to, e.g., scattering to high- k exciton states), a pure dephasing rate, or a combination of both, as only their sum enters in the equation of the coherent fraction. The result also shows that although the impinging laser is very wide in energy, it is possible to prepare the polariton state in a largely tunable range, from almost entirely upper polaritonic (at least for short times) to almost entirely lower polaritonic (also the state at long times), passing by purely photonic and/or excitonic, these two states constantly oscillating between each other.

With such an accurate command of the system, we are able to time precisely the arrival of a second pulse and perform a comprehensive coherent control on the coupled dynamics. For a coupling of the laser to the cavity only, this would be achieved for most operations by sending the control pulse when the cavity field is empty and the state is fully excitonic. Injecting a second fully photonic pulse in optical (antioptical) phase with the exciton, for instance, creates an UP (LP). It is convenient to represent such a dynamics with the joint photon and exciton fields' complex phases, as shown in Fig. 3(a) for a sequence of basic operations through pulsed excitation that bring the system from (i) the vacuum and (vi) back passing by a coherent state of (ii) photons, (iii) UPs, (iv) excitons, and (v) LPs. The photon and exciton states are defined as such right after the pulse only since, not being eigenstates, they enter the oscillating regime. In the rotating frame of the bare modes at frequency ω_0 , the light-matter dynamics is a simple oscillation along the radius with a combined offset of $\pi/2$ both in time and optical phase: the cavity oscillates horizontally while the exciton oscillates vertically and when one reaches its maximum, the other is at the origin. In contrast, the LP and UP do not oscillate radially but circularly, since they are free modes that subtract and add, respectively, their free energy to that of the rotating frame. An animation of this dynamics is given in the Supplemental Material [45].

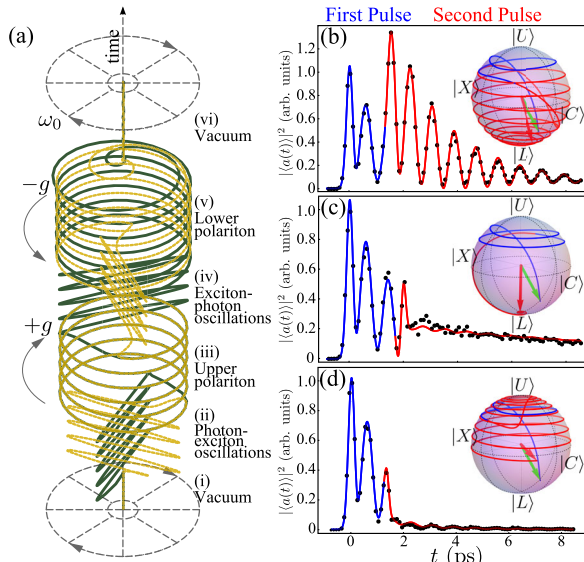


FIG. 3 (color online). (a) Succession of various dynamical evolutions in time of the photon (dashed yellow) and the exciton (green) complex amplitudes (here with fixed modulus) that can be passed from one to the other with an appropriate pulse excitation. Normal mode-coupling features a $\pi/2$ dephasing in both time and optical phase and oscillate radially while polaritons oscillate circularly. Furthermore, they oscillate jointly and with π out of phase with each other and against the rotating frame in the case of a UP (LP). (b)–(d) Experimental realization (points) and theoretical fit (solid curve) of three two-pulses excitation, showing (b) amplification, (c) transition from an exciton-photon Rabi oscillation to a LP, and (d) field annihilation.

In the actual experiment, where the laser couples to both fields, one merely needs to correct for the corresponding proportions but the concept is otherwise the same. A first pulse triggers the Rabi oscillations, since our pulse is broad in energy and always initiates a dominant photon or exciton fraction. However, with a second pulse, although still broad, we can refine the state by providing the complementary of the sought target. Figure 3(b) shows a simple case of Rabi amplification, where the same cycle is restarted by the pulse. Figure 3(c) shows the case where a bare state is transformed into a LP, therefore switching off the oscillations. There is no fundamental difficulty in sending more than two pulses and, in principle, one can prepare any given state right after the pulses. Another case of interest is complete field annihilation, by sending a pulse optically out of phase but in phase with the Rabi oscillations. This produces, by destructive interferences, the vacuum, as shown in Fig. 3(d). All these cases demonstrate the possibility to do coherent control of the strong light-matter coupling dynamics. Here too the theory still provides an essentially perfect agreement to the data. Similar prospects at the single-particle level would perform genuine quantum information processing, but this lies beyond the scope of this work.

In conclusion, we showed the tremendous control that can be obtained on the light-matter coupling in microcavities, for which we reported the first imaging of its spatiotemporal evolution thanks to our femtosecond holographic multiple-pulse excitation. This allowed us to spell out with a precision never achieved before for polaritons both the excitation scheme and the various components involved in the dynamics (dephasing, reservoirs, etc.). We demonstrated the reservoir-induced lifetime enhancement recently proposed [54] and performed coherent control on the polariton state. Such results are a milestone to turn these systems into devices, with future prospects such as optical gates or their single-particle counterpart now clearly in sight. Immediate extensions suggested by this work are—beyond getting to the single-particle limit—to couple to the spatial degree of freedom with packets imparted with momentum or diffusing, and involve nonlinearities at higher pumpings.

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