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Ultrafast non-local control of spontaneous emission

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The radiative interaction of solid-state emitters with cavity fields is the basis of semiconductor microcavity lasers and cavity quantum electrodynamics (CQED) systems. Its control in real time would open new avenues for the generation of non-classical light states, the control of entanglement and the modulation of lasers. However, unlike atomic CQED or circuit quantum electrodynamics, the real-time control of radiative processes has not yet been achieved in semiconductors because of the ultrafast timescales involved. Here we propose an ultrafast non-local moulding of the vacuum field in a coupled-cavity system as an approach to the control of radiative processes and demonstrate the dynamic control of the spontaneous emission (SE) of quantum dots (QDs) in a photonic crystal (PhC) cavity on a ~200 ps timescale, much faster than their natural SE lifetimes.

When a two-level emitter in the excited state interacts with the vacuum field of a cavity mode, the evolution of the system is determined by the interplay of the coupling rate $g$, interaction time $T$ and cavity-loss rate $\kappa$. The control of one or more of these parameters in real time allows optimizing the interaction for the desired application, resulting, for example, in entangled emitter–photon states, or in single-photon states with an optimized waveform for quantum networking applications. In atomic CQED systems, the control of the radiative interaction is realized at microwave frequencies by varying the interaction time $T$ in the range of tens of microseconds, and at optical frequencies by adiabatic passage techniques, which effectively allow the shaping of the coupling rate $g$ on timescales of 100 ns. In superconducting circuit quantum electrodynamics, the cavity-loss rate $\kappa$ has been changed on a timescale of 100 ns by the electrical control of circuit elements. However, in semiconductor systems based, for example, on QDs in PhC cavities, these approaches are difficult to implement because of the different energy-level structures and the faster cavity loss and emitter decoherence rate (typically subnanoseconds). The ideal control method for semiconductor nanocavities would allow the ultrafast manipulation of the coupling rate $g$ and/or of the cavity-loss rate $\kappa$, without directly affecting the population and the phase of the emitter. Although various methods for the control of semiconductor CQED systems have been demonstrated, for example by tuning the emitter or cavity frequency using electric field, strain or nanomechanical deformation, none of these has been shown to provide the control of radiative processes on the required subnanosecond timescales. The fast wavelength-detuning techniques theoretically proposed in Johnson et al. and Thyresson et al. are extremely challenging to implement without directly perturbing the emitter’s evolution, and are intrinsically associated with a wavelength chirp. Here we propose an approach that enables the non-local and ultrafast control of the coupling rate and/or the cavity-loss rate in a solid-state system. We show its implementation in semiconductor QDs weakly coupled to a PhC coupled-cavity system, which demonstrates, for the first time, the ultrafast control of SE dynamics at optical frequencies with a temporal resolution of about 200 ps, which can be reduced further to the few picoseconds range.

Our approach is based on ultrafast moulding of the vacuum field, and thereby the coupling rate and cavity loss, seen by a dipole emitter sitting in a ‘target’ cavity, by changing the resonant wavelength of an adjacent Fabry–Pérot (FP) cavity coupled to the target cavity through a semitransparent mirror (Fig. 1a). When the two cavities are out of resonance, the modes of the system are approximated well by the modes of uncoupled cavities, where, by design, the periodic FP modes have a lower quality factor (Q factor) compared with the mode of the target cavity. A change in the refractive index of the FP cavity produces a spectral shift of the FP modes and brings one of them into resonance with the
target cavity mode (Fig. 1b). This causes a redistribution of the vacuum field seen by the emitter, with a corresponding increase in the effective mode volume and reduction of the Q factor, and thereby changes the emitter-field coupling rate and the loss rate. If the change of the FP spectrum is produced by an ultrafast laser pulse from the photoexcitation of free carriers, the vacuum field responds within a timescale set by the target–FP coupling rate (typically picoseconds) and the emitter experiences a dynamic modulation of the local density of states (LDOS) during its interaction with the cavity. As the free carriers are injected distant from the target cavity (that is, the control is ‘non-local’), they do not directly perturb the population and phase coherence of the emitter. A simple analysis based on coupled-mode theory (see Methods and Supplementary Methods) shows that the emitter interacts with the coupled mode with a rate $g_1 = \alpha g$, where $\alpha$ is the target-cavity component of the electric field of the coupled mode, and $g$ is the interaction rate between the emitter and the uncoupled target cavity. Although this g modulation is generally applicable to any radiative interaction, we focus here on the modulation of the SE rate of emitters weakly coupled to a cavity to give a first experimental demonstration of the concept. The SE rate $\gamma_1$ into one of the coupled modes, normalized by that in the uncoupled target cavity $\gamma_t$, is given by:

$$\frac{\gamma_1}{\gamma_t} = \frac{g_1}{g} \frac{\kappa_1}{\kappa_1} = |\alpha|^2 \frac{Q_1}{Q_t},$$

where $Q_1(\kappa_1)$ and $Q_t(\kappa_t)$ are the Q factors (loss rates) of the coupled mode and uncoupled target cavity, respectively. The SE rate is affected both by the change in the coupling rate $g$ (term $|\alpha|^2$) and loss rate $\kappa$, which are controlled by the target-FP detuning. Also, a pure $g$ modulation is possible by choosing the Q factor of the FP cavity equal to that of the target cavity.

We implemented this concept using PhC cavities and QDs as emitters. Coupled PhC cavities were investigated previously as examples of photonic molecules \(^{15-17}\), for Q-factor tuning \(^{13,14,18}\) and coupled CQED \(^{19}\). Two double-heterostructure cavities \(^{20}\) are defined by slightly modifying the lattice constant along a W1 PhC waveguide from $d_0$ to $d_1 = 1.03 \times d_0$ (Fig. 1c). In the first series of experiments, the change in the SE rate of QDs in the target cavity was characterized in quasi-static conditions by thermo-optic tuning of the FP cavity. The FP modes shift to longer wavelengths because of heating when the excitation power increases \(^{21}\), which changes the detuning between two cavities (Fig. 2a). When the central FP mode anticrosses the target cavity mode, a decrease in emission intensity is observed and the Q factor decreases by a maximum factor of 2.0 (Fig. 2a–c). Depending on the detuning, the cavity wavelength and the Q factor can be fitted well by the coupled-mode theory (Fig. 2b,c). The microphotoluminescence (μPL) decay times strongly depend on the detuning (Fig. 2d) and are reproduced well by the Master equation model, taking into account the homogeneous broadening at the measurement temperature of 77 K, estimated as 250 μeV (see the Supplementary Information). After deducting the measured effect of emission into the leaky modes of the PhC, a change in the SE rate in the cavity mode by a factor of 2.7 is derived, which is attributed partly to the relative change in the effective Q (by a factor of ~1.34, see the Supplementary Information) and partly to the redistribution of the vacuum field term $|\alpha|^2$ (by a factor of ~2).

In a second set of experiments, the dynamic control of SE was achieved by replacing the thermo-optic tuning with free-carrier injection. In this case, a pulsed laser injects free carriers into the FP cavity. When the initial detuning between the target and FP
cavity is adjusted to 0 nm, two coupled modes are observed at 1,552.0 and 1,552.4 nm. The laser pulse produces a blue shift of the FP mode, which brings it out of resonance with the target cavity, and the SE rate from QDs in the target cavity is enhanced because of the increase in the Q factor and the increased localization of the vacuum field (Fig. 3a–c). This gives rise to a peak at time zero in the three-dimensional map, which lasts until the FP mode relaxes to the initial wavelength through diffusion and the recombination of free carriers. The duration of this pulse of SE (232 ps at full-width half-maximum (FWHM)) is related to the free-carrier lifetime in the FP cavity, and not to the emitter’s lifetime—this shows the possibility of modulating SE at frequencies of several gigahertz, well above the bandwidth limitation related to the lifetime. This dynamic process is simulated by a Master equation model (see the Supplementary Information), which includes the effect of pumping by several QDs via their homogeneous broadening and reproduces the PL temporal dependence very well (Fig. 3c). The observed modulation depth at the peak, $I_{\text{max}}/I_0 = 3.3$, results from the combined effect of the LDOS enhancement, the increase of photon population caused by the Q change and the change in the collection efficiency as a result of the redistribution of the vacuum field.

For the opposite situation of non-zero detuning, the FP and target cavity modes can be transiently brought into resonance (Fig. 3d–f). This produces a sharp dip (246 ps FWHM) in the SE intensity because of the LDOS reduction caused by the vacuum field delocalization and Q-factor decrease, which confirms that the peak in Fig. 3a is not a result of the additional PL produced by the injected carriers. For this case, the measured peak modulation depth is $I_0/I_{\text{max}} = 2.0$. Here the modulation occurs over the entire mode spectrum, and clearly differs from the static modulation of the SE rate using PhC cavities and the dynamic modulation, which may, in principle, be obtained by changing the emitter–cavity detuning with ultrafast control of the cavity wavelength or by Stark tuning of the exciton energy. Indeed, our method directly changes the SE rate by controlling the interaction term and loss rate $g$ and $\kappa$. This allows the chirp of the emitted photons to be minimized. This chirp reduces the coupling efficiency to another cavity and is therefore detrimental to application in quantum-information processing.

To demonstrate further the flexibility of our approach, we show the control of the temporal SE decay profile of the emitters in the target cavity. In this case, both the target and the FP cavity are pumped by the same pulsed laser, with variable delay. The excitation power at the target cavity produces an initial blue shift in the target cavity mode (as seen in the inset of Fig. 4a) caused by free-carrier injection. To avoid the effect of this blue shift we choose delays such that the target cavity wavelength is stabilized. When the initial detuning is zero and the pulse exciting at the FP cavity is delayed by 2.0 ns, a spike is observed in the PL signal collected from the target cavity when the FP mode is brought out of resonance. The timing of the spike can be controlled by the delay, as shown in Fig. 4a for delays of 1.5, 2.0 and 2.5 ns. For the opposite situation in which the static detuning between coupled modes is set to 0.6 nm, the FP mode is transiently brought into resonance so that the expected dip appears (Fig. 4b).

In our demonstration, the temporal resolution is determined by the free-carrier lifetime, which potentially can be reduced down to a few picoseconds by applying an electric field in the control cavity. This would allow shaping of the control-cavity frequency and thereby the LDOS at the target cavity on picosecond timescales by the simple control of the pump–pulse temporal profile.
In the context of CQED, this ultrafast non-local control can be used to engineer the shape of single-photon pulses and for the on–off switching of Rabi oscillations, without directly disturbing the carrier population in the target cavity. Although, for simplicity, the experiments presented here were performed with an ensemble of QDs, the extension to the single-QD/single-photon case is straightforward, as single QDs are easily coupled to PhC cavity modes\(^1\). Although our initial demonstration focused on the control of SE, the mode field also determines the stimulated emission rate. Our method of ultrafast moulding of the vacuum field can therefore be used to change dynamically the Q factor and/or the gain in lasers. The ultrafast Q-factor tuning directly affects the laser output via the cavity-loss term, and it opens the way to Q-switching in nanocavity lasers\(^6\). The modulation of the cavity field, however, changes the modal gain via the confinement factor, and represents a new type of gain switching that does not rely on directly altering the carrier population via current injection.

**Methods**

**Coupled-mode theory.** The system of three coupled oscillators (the emitter, the target and FP cavities) can be described by a non-Hermitian Hamiltonian:\(^7\)

\[
H = H \begin{bmatrix}
\omega_0 & g & 0 \\
g & \omega_0 - \omega_1 - \eta & \eta \\
0 & \eta & \omega_0 - \omega_2 - \kappa_2 & \kappa_2
\end{bmatrix}
\]

where \(\omega_0\), \(\omega_1\) and \(\omega_2\) are the angular frequencies of the emitter, the uncoupled target and the FP cavity modes, respectively, and \(\kappa_1\) and \(\kappa_2\) are the corresponding loss rates. We assume that the emitter couples only to the target cavity with an interaction strength \(g\), where we fix the interaction to the weak coupling regime \(g \ll \kappa_1, \kappa_2\). The coupling rate between the two cavities is \(\eta\). By diagonalizing the \(2 \times 2\) submatrix of the cavities, the normalized mode functions \(E_i(x)(t)\) of the coupled modes are calculated on the basis of the isolated cavity modes \(E_i(x)\) and \(E_P(x)\),

\[
E_i(x)(t) = a_i \tilde{E}_i(x) + \beta_i \tilde{E}_P(x)\text{ with } a_i = \beta_i = 0, \text{ and } a_i = \beta_i = \beta_0.\]

In the basis of the coupled modes \(E_i(x)(t)\), the Hamiltonian is given by:

\[
H = H_0 + H_1,
\]

\[
H_1 = \begin{bmatrix}
\beta_0 & \tilde{E}_0 & \tilde{E}_1 & \tilde{E}_2 \\
\tilde{E}_0 & \omega_0 & \beta_0 & 0 \\
\tilde{E}_1 & \beta_0 & \omega_1 - \omega_0 - \kappa_1 & \kappa_1 \\
\tilde{E}_2 & 0 & \kappa_1 & \omega_2 - \omega_0 - \kappa_2
\end{bmatrix}
\]

where \(\tilde{E}_0 = \tilde{E}_1 + \kappa_1\) are the complex eigenvalues of the coupled modes. The effective dipole-coupled-mode interaction rates \(g = \kappa_2\) and the interaction rates \(g = \beta_0\) and \(g = \beta_0\) and \(\tilde{E}_2 = \beta_0\). In the basis of the uncoupled modes \(E_i(x)\) and \(E_P(x)\),

\[
\tilde{E}_i(x)(t) = a_i \tilde{E}_i(x) + \beta_i \tilde{E}_P(x)\text{ with } a_i = \beta_i = 0, \text{ and } a_i = \beta_i = \beta_0.\]

The measurement was performed at 77 K using a confocal laser, at different delays, with energies of 20 and 200 \(\mu\)J cm\(^{-2}\) per pulse, respectively. The fine adjustment of the wavelength detuning between two cavities was achieved by coupling a CW lasing beam at 780 nm on the FP cavity.

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Author contributions
A.F. proposed the experiment and led the project. C.-Y.J., R.J., A.F. performed the measurements. R.J. developed the theory. C.-Y.J., M.Y.S. and L.M. fabricated the devices. C.-Y.J., M.Y.S. and L.M. performed the optical simulations. P.J.V. grew the sample. C.-Y.J. designed and fabricated the devices. C.-Y.J., M.Y.S. and L.M. proposed the experiment and led the project. C.-Y.J., M.Y.S. and L.M. performed the fabrication and experiments, to Z. Zhou, D. Sahin, F. M. Pagliano, C. P. Dietrich, E. J. Gelak, E. Smalbrugge, T. de Vries, M. van Vlokhoven, J. M. van Ruijven and P. A. M. Nouwens for technical support, to V. Savona for useful discussions on the theoretical aspects and to P. M. Koenraad and E. Pelucchi for a critical reading of the manuscript. This research is supported financially by NanoNexNL, a micro and nanotechnology program of the Dutch Ministry of Economic Affairs, Agriculture and Innovation (EL&I) and 130 partners, the Dutch Technology Foundation STW, Applied Science Division of NWO, the Technology Program of the Ministry of Economic Affairs under project No. 10380 and the FOM project No. 09PR2675. One of the authors (A.F.) dedicates this work to the memory of E. Rosencher.

Additional information
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Competing financial interests
The authors declare no competing financial interests.