How fast electrons and photons mix: sub-cycle switching of intersubband cavity polaritons

R Huber, A A Anappara, G Günter, A Sell, G Biasiol, L Sorba, A Tredicucci, S De Liberato, C Ciuti, and A Leitenstorfer

1 Department of Physics, University of Konstanz, 78464 Konstanz, Germany
2 Laboratorio NEST, CNR-INFM and Scuola Normale Superiore, 56127 Pisa, Italy
3 Laboratorio Nazionale TASC CNR-INFM, 34012 Trieste, Italy
4 Laboratoire Matiereaux et Phenomenes Quantiques, CNRS and Universite Paris Diderot-Paris 7; Laboratoire Pierre Aigrain, ENS, 75005 Paris, France

E-mail: rupert.huber@uni-konstanz.de

Abstract. Intersubband cavity polaritons in a quantum well waveguide structure are optically generated within less than one cycle of light by a femtosecond near-infrared pulse. Mid-infrared probe transients trace the non-adiabatic switch-on of ultrastrong light-matter coupling and the conversion of bare photons into cavity polaritons directly in the time domain.

1. Introduction
Photonic microcavities have been exploited to tailor the interaction of light with material excitations. In the strong coupling regime, a photon may be absorbed and spontaneously reemitted by an elementary excitation many times before dissipation becomes effective. This process gives rise to new eigenstates of mixed light-matter character, so-called cavity polaritons. The characteristic energy anticrossing, known as vacuum-field Rabi splitting $2\hbar \Omega_R$, is a direct measure of the strength of light-matter interaction [1]. Recently intersubband resonances of semiconductor quantum wells (QW) hybridized with the mid-infrared photon mode of a planar waveguide have entered a new regime of ultrastrong interaction, where $\Omega_R$ amounts to a significant fraction of the bare eigenfrequencies $\omega_{12}$ themselves [2-5]. The resulting squeezed two-mode quantum vacuum is expected to give rise to a variety of novel quantum electrodynamical (QED) effects [6]. In particular, ultrafast switching of the Rabi frequency $\Omega_R$ has been predicted to release correlated photon pairs out of the vacuum, reminiscent of the intriguing, yet unobserved dynamic Casimir effect [6].

Up to now, however, non-adiabatic phenomena – classical or QED in nature – remained an academic curiosity since there has been no laboratory capable of controlling light-matter interaction on a sub-cycle time scale. Here, we report an all-optical pump – terahertz probe scheme for the first implementation of femtosecond control of ultrastrongly coupled cavity polaritons [7].

2. Multi-THz waveguide with semiconductor quantum wells
The sample contains 50 identical, undoped GaAs QWs separated by Al$_{0.33}$Ga$_{0.67}$As barriers. The electronic wave functions are quantized along the growth direction forming subbands (Fig. 1). These are unpopulated in thermal equilibrium. Radiative transitions between subbands of quantum number $n = 1$ and $n = 2$ are activated if optical excitation promotes electrons from the valence into the...
conduction band. The intersubband resonance features a narrow absorption line centered about a photon energy of $\hbar \omega = 113$ meV (wavelength $\lambda = 11 \, \mu m$) and a strong dipole moment oriented along the growth direction. The multi-QW structure is designed as a planar step index waveguide for mid-infrared light [2]. Radiation is confined between a top-cladding (Al$_{0.33}$Ga$_{0.67}$As)-air interface ($n_{\text{air}} = 1$, $n_{\text{QW}} = 3.1$) on one side and a low refractive index AlAs layer ($n_{\text{AlAs}} = 2.9$) on the other. The effective thickness of the entire waveguide is chosen to be $\lambda/2$ at an internal angle of $\theta = 65^\circ$. Photon modes with electric field components in growth direction (TM polarization) may resonantly couple to intersubband transitions provided the subbands are populated.

Figure 1. Schematic of the femtosecond optical switch: 50 undoped GaAs quantum wells of a thickness of 9 nm each, separated by Al$_{0.33}$Ga$_{0.67}$As barriers are embedded into a planar waveguide based on total internal reflection. Intersubband transitions are activated by near-infrared 12-fs control pulses populating level $|1\rangle$. TM-polarized multi-THz transients guided through the prism-shaped substrate are reflected off the waveguide to probe the ultrafast buildup of intersubband cavity polaritons.

3. All-optical control of light-matter coupling: from zero to ultrastrong

We employ a low-noise Ti:sapphire laser system generating amplified 12-fs pulses (central photon energy: 1.55 eV) to photoinject electrons from the valence band into the lowest conduction subband of the QWs (Fig. 1), activating the intersubband oscillator. Since the inverse frequency of the latter transition amounts to 37 fs, switching occurs within less than half a cycle of light. The subsequent ultrafast dynamics of the non-equilibrium cavity at room temperature is traced by multi-terahertz (THz) spectroscopy [8]: A second part of the laser output generates phase-locked THz pulses covering the spectral window from 80 meV to 150 meV by optical rectification in a 50-µm-thick GaSe emitter [9]. TM-polarized field transients are coupled through the prism-shaped substrate and internally reflected off the photexcited area of the waveguide under incidence angles around $\theta = 65^\circ$ (Fig. 1). The pulse front of the near-infrared pump is tilted to match the geometry of the THz phase surfaces. The oscillating electric field of the reflected THz transient is resolved in the time domain via phase-matched electro-optic sampling [9]. Fourier transformation provides amplitude and phase spectra in the mid infrared. The eigenmodes of the cavity are identified via their characteristic minima in the amplitude reflectivity [2].

In a first step, we demonstrate that light-matter coupling is continuously tunable via the control fluence $\Phi$. The spectra of Fig. 2 are recorded at a fixed delay $t_0 = 20$ ps between the near-infrared control and the multi-THz probe pulse. In equilibrium ($\Phi = 0$), a single reflectance minimum at $\hbar \omega_c = 113$ meV (top curve) attests to the sole resonance of the unexcited cavity, the bare photon mode. For $\Phi > 0.05 \times \Phi_0$, $\Omega_R$ exceeds the widths (FWHM ~ 5 meV) of intersubband and cavity resonances and two cavity polariton branches are clearly discernible. At this point, the system crosses from the weak to the strong coupling regime. Further increase of the fluence enhances the separation of the minima to as much as 50 meV (see e.g. bottom curve of Fig. 3), corresponding to a fraction of 44% of the bare photon frequency. The apparent mode separation is not identical with the vacuum Rabi splitting at the anti-crossing point [2]. Only a quantitative simulation of the energy position of the polariton dips allows for extraction of $\Omega_R$. For a correct description of our data, the theory has to go beyond the rotating wave approximation and include antiresonant terms in the light-matter Hamiltonian. These contributions describe the simultaneous creation or annihilation of two excitations with opposite in-
plane wave vector $k$ and give rise to a two-mode squeezed quantum vacuum [6]. By comparison with this theory we determine a maximum splitting $2\Omega_R = 0.18 \times \omega_{12}$ for our experiments. This value is comparable to the record achieved in doped structures [3,4] and clearly fulfills the criteria for ultrastrong coupling [6].

Figure 2. All-optical in-situ tuning of light-matter interaction. THz reflectance spectra measured at room temperature for various fluences $\Phi$ (vertically offset) of the control pulse ($t_0 = 20$ ps). Minima indicate eigenmodes of the system. For $\Phi = 0$, only the bare photon mode is observed at $\hbar \omega_c = 113$ meV; both branches of the intersubband cavity polaritons are discernible for $\Phi \geq 0.05 \Phi_0$, where $\Phi_0 = 0.1$ mJ/cm$^2$.

4. Non-adiabatic switch-on of intersubband cavity polaritons

The central question is: How rapidly may ultrastrong coupling be activated? In order to address this issue, we repeat the experiments for various delay times $t_D$ between near-infrared pump and multi-THz probe pulses. The results are illustrated in Fig. 3a: For $t_D \leq -50$ fs the spectra are dominated by a single dip located at 113 meV which we may unequivocally assign to the photonic waveguide mode. Photoinjection of electrons into the lower subband induces dramatic changes of the spectra of order unity. The initial bare photon eigenstate is replaced, on a ten femtosecond scale, by two ultrastrongly coupled cavity polariton modes appearing simultaneously at energies of 94 meV and 143 meV, respectively (arrows in Fig. 3). Most remarkably, the new resonances do not develop by gradual bifurcation out of the bare cavity mode as in Fig. 2. In contrast, switching occurs discontinuously once subband $|1\rangle$ is populated.

Femtosecond activation of the light-matter Hamiltonian is predicted to give rise to yet unexplored effects on pre-existing photon states and the quantum vacuum itself [6]. This class of phenomena is best studied directly in the time domain (see Fig. 3b): When a few-cycle probe transient impinges on the unexcited modulator, part of its energy is reflected off the cavity surface. A second portion is evanescently coupled into the resonator, prepares a coherent photon state, and gets reemitted subsequently. This dynamics is encoded in a characteristic twin-pulse structure of the reflected transient (see curve (ii)). The most intriguing situation is realized if we turn light-matter coupling on while a coherent state of bare photons is still present inside the cavity: The control pulse (vertical arrow in curve (iii)) abruptly alters $\Omega_R$ during the free cavity decay. Remarkably, the emission of bare photons is interrupted on a time scale shorter than half an oscillation cycle of light – a compelling proof of non-adiabaticity. The subsequent field trace exhibits a characteristic two-mode beating (inset in Fig. 3b), the hallmark of coherent oscillations of both polariton branches. Thus we do not only control the eigenstates of the microcavity, but effectively convert a coherent photon population into ultrastrongly coupled cavity polaritons beating at the splitting frequency. We suggest the term “perturbed cavity decay” (PCD) to describe this novel sub-cycle phenomenon. A quantitative theory including anti-resonant terms of the light-matter Hamiltonian as well as the extremely non-equilibrium carrier dynamics is currently under way.
Figure 3. Two-dimensional optical pump – THz probe data. a Amplitude reflectivity spectra at $T_L = 300$ K for various delay times $t_D$ (control fluence: $\Phi = 0.1$ mJ/cm$^2$). The arrows serve as guides to the eye for the position of the cavity and polariton resonances, respectively. b Nonadiabatic conversion of bare photons into cavity polaritons, in the time domain: (i) incident THz transient, (ii) reflected transient without control pulse. The twin pulse structure is due to radiation directly reflected off the cavity surface (first burst) and bare photons reemitted from the cavity (second pulse). (iii) The near-infrared control pulse (vertical arrow) alters the cavity emission on a sub-cycle scale: The exponential bare cavity decay is replaced by the characteristic polariton beating (insert, magnified by factor 8).

5. Conclusions
We have demonstrated the first non-adiabatic switching scheme of cavity-polaritons, reaching the regime of ultrastrong light-matter coupling on a sub-cycle scale. The experiments provide a benchmark for latest theories in the ultrastrong coupling regime, point out a viable route towards novel QED phenomena such as the observation of Casimir-type vacuum radiation, and demonstrate a room-temperature optical switching device at the ultimate speed.

[1] Weisbuch C et al. 1992 Phys. Rev. Lett. 69 3314
[2] Dini D et al. 2003 Phys. Rev. Lett. 90 116401
[3] Anappara A A et al. 2009 Phys. Rev. B Rapid 79 201303
[4] Todorov Y et al. 2009 Phys. Rev. Lett. 102 186402
[5] Anappara A A et al. 2005 Appl. Phys. Lett. 87 051105; 2006 Appl. Phys. Lett. 89 171109
[6] Ciuti C, Bastard G, and Carusotto I 2005 Phys. Rev. B 72 115303; De Liberato S et al. 2007 Phys. Rev. Lett. 98 103602
[7] Günter G et al. 2009 Nature 458 178
[8] Huber R et al. 2001 Nature 414 286; 2005 Phys. Rev. Lett. 94 027401; Kübler C et al. 2007 Phys. Rev. Lett. 99 116401
[9] Kübler C et al. 2005 Semicond. Sci. Technol. 20 128 and references therein