

Non-adiabatic switch-off and subcycle nonlinearities of deep-strong light-matter coupling

C. Lange^{1,2}, J. Mornhinweg¹, M. Halbhuber¹, V. Zeller¹,
C. Ciuti³, D. Bougeard¹, and R. Huber¹

¹Department of Physics, University of Regensburg, 93040 Regensburg, Germany

²Department of Physics, TU Dortmund University, 44227 Dortmund, Germany

³Université de Paris, Laboratoire Matériaux et Phénomènes Quantiques, CNRS, F-75013, Paris, France

E-mail: christoph.lange@tu-dortmund.de

Dressing electrons with virtual photons of an optical cavity can create new states of matter. When the rate of energy exchange between the light and the matter mode – the vacuum Rabi frequency, Ω_R – approaches the carrier frequency of light ω_0 , the regime of ultrastrong coupling is attained [1,2]. Here, anti-resonant interactions drive two-particle creation and annihilation events on subcycle time scales and the ground state becomes populated by virtual photon pairs. As a result, novel effects of cavity quantum electrodynamics (c-QED) arise, such as the vacuum Bloch-Siegert shift [3], modified electronic transport [4], and cavity-controlled changes in chemical reactions [5]. For deep-strong coupling [6,7], Ω_R even exceeds ω_0 , leading to unconventional c-QED phenomena including cavity-mediated superconductivity [8]. While some properties of this regime may be explored under equilibrium conditions, controlling the coupling mechanism on strongly subcycle scales is expected to reveal fundamentally new quantum phenomena.

Here, we explore the subcycle dynamics that arises when light-matter interaction of a deep-strongly coupled system is switched off extremely non-adiabatically [7]. We employ custom-tailored THz antennas coupled to the cyclotron resonance (CR) of two-dimensional electron gases hosted in semiconductor quantum well structures. As a result, new eigenstates called cavity polaritons form, evidenced by characteristic minima of the transmission which are separated by $2\Omega_R$ when the CR is tuned to the optical mode. In our most strongly coupled structure, this separation of the lower (LP) and upper polariton (UP) branch corresponds to a normalized coupling strength of $\Omega_R/\omega_c = 1.3$ and a ground state population of 0.32 virtual photons. Switching of light-matter coupling is achieved by introducing an $\text{In}_{0.55}\text{Ga}_{0.45}\text{As}$ semiconductor patch in the gap region of the resonator, where the THz field enhancement is maximal. Near-infrared femtosecond photoexcitation of this patch forms a charge carrier plasma which screens the cavity mode and diminishes its amplitude to less than 5% of the value prior to excitation, completely collapsing light-matter coupling, and the polariton branches. In this extraordinary setting, we perform subcycle time-domain spectroscopy to extract the linear response function and time-dependent transmission spectra as a function of the pump-probe delay time. Intriguingly, the collapse of the LP is accompanied by trailing oscillations of the transmission with frequency components exceeding the LP frequency several times. Our quantum model including anti-resonant interaction terms links these high-frequency dynamics to an abrupt, subcycle collapse of Ω_R within just 5% of the cycle duration of the LP. Moreover, the calculations highlight a path towards converting the virtual ground state population to real excitations [7,9] in similarity to Unruh-Hawking radiation of black holes. Finally, we employ two-dimensional THz spectroscopy which reveals high-order nonlinearities including up to eight-wave mixing, and nonlinear inter-polariton correlations. Our custom theory links these effects to dynamical mixing of the equilibrium eigenstates of the structure in a setting of non-perturbative excitation. In both concepts, subcycle excitation acts as a new control parameter for light-matter hybridization, setting the stage for novel effects of c-QED.

- [1] A. Günter et al. *Sub-cycle switch-on of ultrastrong light-matter interaction*, Nature **458**, 178 (2009).
- [2] G. Scalari et al. *Ultrastrong coupling of the cyclotron transition of a 2D electron gas to a THz metamaterial*, Science **335**, 1323 (2012).
- [3] X. Li et al. *Vacuum Bloch-Siegert shift in Landau polaritons with ultra-high cooperativity*, Nat. Photon. **12**, 324 (2018).
- [4] G. L. Paravicini-Bagliani et al. *Magneto-transport controlled by Landau polariton states*, Nat. Phys. **15**, 186 (2019).
- [5] A. Thomas et al. *Tilting a ground-state reactivity landscape by vibrational strong coupling*, Science **363**, 615 (2019).
- [6] A. Bayer et al. *Terahertz Light-Matter Interaction beyond Unity Coupling Strength*, Nano Lett. **17**, 6340 (2017).
- [7] M. Halbhuber et al. *Non-adiabatic stripping of a cavity field from electrons in the deep-strong coupling regime*, Nature Photonics **14**, 675 (2020).
- [8] F. Schlawin, A. Cavalleri, D. Jaksch, *Cavity-mediated electron-photon superconductivity*, Phys. Rev. Lett. **122**, 133602 (2019).
- [9] S. De Liberato, C. Ciuti, I. Carusotto, *Quantum Vacuum Radiation Spectra from a Semiconductor Microcavity with a Time-Modulated Vacuum Rabi Frequency*, Phys. Rev. Lett. **98**, 103602 (2007).
- [10] J. Mornhinweg et al., *Tailored subcycle nonlinearities of ultrastrong light-matter coupling*, arXiv:2011.14635 (2020).