Direct measurement of light–polarization beating inside a microcavity: supplementary material

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In this supplementary information (SI) to "Direct measurement of light–matter energy exchange inside a microcavity," <http://dx.doi.org/10.1364/optica.1.000276>, we briefly present details on the experimental setup and discuss the impact of excitation-induced dephasing (EID) effects on the computed spectra. Additionally, we present a direct experiment–theory comparison of the differential reflectivity. © 2014 Optical Society of America

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1. EXPERIMENTAL DETAILS

In our investigations, we carry out time-resolved optical and THz measurements to observe the ultrafast nonlinear dynamics of the exciton–polariton oscillations in a QW microcavity driven by strong single-cycle THz pulses.

Fig. S1. Experimental setup. Experimental scheme of time-resolved strong-THz and weak-optical excitations. See text for more details.

Supplementary Figure S1 sketches the experimental setup. The light source is a 1-kHz Ti:sapphire regenerative amplifier

(pulse duration: 80 fs; central wavelength: 800 nm; pulse energy: 1 mJ). Using a small portion of the pulse energy (10 μ J), we produce weak 830-nm, 100-fs optical pulses by white-light continuum generation in a 1-mm quartz crystal with a bandpass filter (central wavelength: 830 nm; bandwidth: 10 nm). The major portion of the pulse energy is used to generate THz pulses by optical rectification in a 1-mm $\langle 110 \rangle$ ZnTe crystal. The crystal is excited by a collimated laser beam of 3 mm in diameter. The THz radiation is focused on the QW microcavity with a spot size of 1.5 mm in diameter. The THz-field amplitude reaches 10 kV/cm at the focus. Using the optical pulses, we measure the reflectivity spectra of the lower and higher exciton–polariton (LEP and HEP) modes in the QW microcavity while varying the relative time delay between the optical-pump and THz-reset pulses ($\Delta t > 0$, when the THz pulse arrives at the sample after the optical pulse does). The optical spectra are acquired by an imaging spectrometer with a CCD camera (spectral resolution: 0.01 nm). The QW microcavity sample (NMC66) consists of 10 InGaAs QWs in a $11 \lambda/2$ -microcavity with distributed Bragg reflectors (DBRs) designed for 99.4% reflectivity. Exploiting the wedged cavity, we tune the cavity resonance by scanning the optical beam across the sample. The sample temperature is maintained at 5 K in a L-He cryostat.

We perform the experiments controlling three parameters: optical-pump–THz-reset time delay (Δt), THz-field amplitude (E_{THz}), and cavity detuning (δ_c). Supplementary Figure S2a illustrates the energy-level structure of the Λ system consisting of

Fig. S2. Nonlinear optical effects of exciton-polaritons in a QW microcavity excited by strong single-cycle THz pulses. (a) THz power spectrum and the energy-level structure of the exciton-polariton Λ -type three-level system (the resonance frequencies of the LEP-to- $2p$ and the HEP-to- $2p$ transitions at zero cavity detuning, $\delta_c = 0$, are 2.5 THz and 1.1 THz, respectively), (b) THz-induced bleaching of the exciton-polariton modes without reset (gray-solid) and with reset (red-solid) at $\delta_c = 0$ at $\Delta t = 0.5$ ps in the presence of a THz field, $E_{\text{THz}} = 10$ kV/cm.

HEP, LEP, and $2p$ -exciton state [1], where the resonance frequencies of the LEP-to- $2p$ and the HEP-to- $2p$ transitions at $\delta_c = 0$ are 2.5 THz and 1.1 THz, respectively. The resonance of the HEP-to- $2p$ transition is in the middle of the THz spectrum while the THz radiation is off-resonant with respect to the LEP-to- $2p$ transition. The nonlinear THz effects are, however, remarkably strong even for the off-resonant transition at the high THz intensity: both LEP and HEP modes are almost wiped out at $\Delta t = 0.5$ ps in the presence of a THz field, $E_{\text{THz}} = 10$ kV/cm, as shown in Supplementary Figure S2 b. The THz reset of QW polarization is nearly complete in this extreme-nonlinear-optics regime.

2. THEORETICAL APPROACH

In our work presented in the main manuscript, we study optical-pump and THz-reset response in a microcavity. Such a configuration constitutes a complicated optical system consisting of alternating layers of different materials. The propagation of THz and optical fields is computed by solving the wave equation coupled to matter polarization (P) and currents (J) [2, 3]. The transmission, reflection, and absorption is solved self-consistently by solving P microscopically from the semiconductor Bloch equations (SBEs) [4].

Our many-body theory is based on the system Hamiltonian that includes noninteracting electron and hole contributions,

the Coulomb interaction between charge carriers, the carrier-phonon interaction, as well as the light-matter interaction [5, 6]. Using the Heisenberg equations-of-motion approach [4] for the relevant operator combinations, we truncate the arising operator hierarchy systematically by applying the cluster-expansion approach [7]. The approach leads to the Maxwell-semiconductor Bloch equations [4, 7] including THz contributions [5]:

$$\begin{aligned}
 i\hbar \frac{\partial}{\partial t} P_{\mathbf{k}} &= \left(\epsilon_{\mathbf{k}} - \mathbf{j}_{\mathbf{k}} \cdot \mathbf{A}_{\text{THz}} + \frac{|e|^2}{2\mu} \mathbf{A}_{\text{THz}}^2 \right) P_{\mathbf{k}} \\
 &\quad - \left(1 - f_{\mathbf{k}}^e - f_{\mathbf{k}}^h \right) \left(d_{cv} E_{\text{opt}} + \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'} P_{\mathbf{k}'} \right) \\
 &\quad + \Gamma_{\mathbf{k},\text{phon}}^{vc} + \Gamma_{\mathbf{k},\text{Coul}}^{vc}, \\
 i\hbar \frac{\partial}{\partial t} f_{\mathbf{k}}^e &= 2 \text{Im} \left[P_{\mathbf{k}} \left(d_{cv} E_{\text{opt}} + \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'} P_{\mathbf{k}'} \right)^* \right. \\
 &\quad \left. + \Gamma_{\mathbf{k},\text{phon}}^{cc} + \Gamma_{\mathbf{k},\text{Coul}}^{cc} \right], \\
 i\hbar \frac{\partial}{\partial t} f_{\mathbf{k}}^h &= 2 \text{Im} \left[P_{\mathbf{k}} \left(d_{cv} E_{\text{opt}} + \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'} P_{\mathbf{k}'} \right)^* \right. \\
 &\quad \left. - \Gamma_{\mathbf{k},\text{phon}}^{vv} - \Gamma_{\mathbf{k},\text{Coul}}^{vv} \right]. \tag{1}
 \end{aligned}$$

Here, $P_{\mathbf{k}}$ is the microscopic optical polarization at carrier momentum $\hbar\mathbf{k}$, and $f_{\mathbf{k}}^e$ and $f_{\mathbf{k}}^h$ are the corresponding electron and hole densities, respectively. Structurally, the optical Rabi frequency $d_{cv} E_{\text{opt}}$ generates the microscopic polarization whereas the Coulomb interaction $V_{\mathbf{k}-\mathbf{k}'}$ renormalizes it. In our experiments, we use a moderate optical pump resulting in only low densities ($f_{\mathbf{k}}^{e/h} \ll 1$) such that the phase-filling factor $(1 - f_{\mathbf{k}}^e - f_{\mathbf{k}}^h)$ is modified only minutely. Therefore, the time evolution of the carrier densities may be neglected for the dilute excitation conditions reached in our experiment. The renormalized kinetic electron-hole energy is given by $\epsilon_{\mathbf{k}}$. The THz vector potential $\mathbf{A}_{\text{THz}}$ couples to the current-matrix element $\mathbf{j}_{\mathbf{k}} = -|e|\hbar\mathbf{k}/\mu$ where $\mu = m_e m_h / (m_e + m_h)$ is the reduced effective mass of an electron-hole pair with effective masses $m_e = 0.0665m_0$ for electrons and $m_h = 0.235m_0$ for holes, where m_0 is the bare electron mass. This THz contribution also induces ponderomotive contributions [8] scaling with electron charge e and reduced effective mass μ [5, 9]. Scattering of charge carriers among each other and with phonons leads to energy renormalizations and dephasing of the polarization. Such processes are expressed via higher-order correlations $\Gamma_{\mathbf{k},\text{Coul}}^{vc}$ and $\Gamma_{\mathbf{k},\text{phon}}^{vc}$ where Coulomb and phonon interactions account for the correct momentum exchange in the scattering process. In linear response, both yield $\Gamma_{\mathbf{k},\text{Coul}}^{vc} + \Gamma_{\mathbf{k},\text{phon}}^{vc} = \sum_{\mathbf{k}'} \Gamma_{\mathbf{k},\mathbf{k}'} P_{\mathbf{k}'}$ describing diffusive scattering of $P_{\mathbf{k}}$ into $P_{\mathbf{k}'}$ [10, 11]. Technical details may be found in, e.g., Refs. [7, 10]. The impact of these scattering processes on polarization dephasing and thus on the resulting spectra is discussed in Sec. 3.

We solve the SBEs self-consistently together with Maxwell's wave equation [12],

$$\left(\frac{\partial^2}{\partial z^2} - \frac{n(z)^2}{c^2} \frac{\partial^2}{\partial t^2} \right) E_{\text{opt}}(z, t) = \mu_0 \delta(z) \frac{\partial^2 P}{\partial t^2} \tag{2}$$

having refractive-index profile $n(z)$, the vacuum speed of light c , and the vacuum permeability μ_0 , to describe the light propagation along the z -direction within the particular sample [13]. The self-consistent coupling follows from the macroscopic polarization P which is a sum of the microscopic polarizations $P_{\mathbf{k}}$,

weighted by the dipole-matrix element d_{cv} :

$$P = \frac{d_{cv}}{S} \sum_{\mathbf{k}} P_{\mathbf{k}} + \text{c. c.} \quad (3)$$

where c. c. stands for the complex conjugation and S is the quantization area. This way, we systematically include backcoupling of the excited quantum wells on the electromagnetic fields. In other words, the optical field E_{opt} is changed by the macroscopic polarization P in each quantum well at position z .

The optical field E_{opt} contains both transmitted, $E_T(z, t)$, and reflected, $E_R(z, t)$, field that propagate away from the quantum-well microcavity. The reflection R is identified from the Fourier transform of the reflected field $E_R(\omega)$ to construct the reflection probability $R(\omega) = |E_R(\omega)/E_{\text{in}}(\omega)|^2$ where E_{in} is the Fourier transform of the incoming field. We apply the transfer-matrix method [2, 3] to compute the propagation of both THz and optical fields in each quantum well of our multiple-QW structure such that the final response is constructed of the combined interplay of all 10 quantum wells embedded in our microcavity sample. Because THz fields and optical fields are located in considerably different frequency ranges, the THz field is only weakly influenced by the optical cavity, unlike the optical field. The dynamics of the polarization follows from the SBEs introduced in Eq. (1). In combination with the Maxwell-SBEs used to systematically couple the classical light field to the macroscopic polarization, we eventually obtain a closed set of equations which may be solved self-consistently thus allowing a realistic simulation of the pump-reflection spectra presented in the main manuscript.

3. EXCITATION-INDUCED DEPHASING

As a result of electron-hole excitations in the system, scattering due to Coulomb and phonon interactions leads to state-dependent broadening of excitonic resonances for elevated densities. This effect is often referred to as *excitation-induced dephasing* (EID) [7, 14–19] where the diffusive character of the scattering process redistributes polarizations and leads to a decay of the polarizations to zero within few picoseconds. Besides the state-dependent broadening where exciton states beyond $1s$ show larger dephasing constants than the $1s$ -exciton resonance, i.e., $\gamma_{\lambda} > \gamma_{1s}$, EID also leads to clustering and shifting of higher states with respect to the $1s$ -exciton energy. In general, the effects of EID depend strongly on the configuration in the system. Therefore, a theoretical investigation with respect to the current many-body configuration opens up the possibility to determine the fraction of excitons to plasma in the system of investigation, among others [20].

In our fully microscopic computations presented in the main manuscript and in Sec. 2 of this SI, the effect of EID is manifest in the fact that both HEP and LEP become broader and the intracavity quantum beats which are visible on the THz-induced transients become diminished. The described effects can clearly be seen by comparing Fig. 4a of the main manuscript with Supplementary Figure S3 where EID effects have not been included. Since these effects lead to broadening and shifting of exciton states, mainly the HEP, being closer to exciton states, will be influenced by EID. This directly explains why the oscillations on the HEP are much more attenuated when EID is taken into account than those on the LEP; compare the computed ΔR spectra with (Fig. 4a of the main manuscript) and without (Supplementary Figure S3) EID effects. Comparing the measurement presented in Fig. 3a of the main manuscript with our fully microscopic

Fig. S3. Computed polariton oscillations in pump-reset protocol, without EID effects. Differential reflectivity $\Delta R = R_0 - R_{\text{reset}}$ as a function of energy and delay between optical pump and THz-reset pulse. Excitation-induced effects have been switched off in this computation, in contrast to Fig. 4a of the main manuscript.

computations including and neglecting EID effects, respectively, we may conclude that excitation of our sample indeed exerts a considerable influence on exciton states and thus the overall line shape.

To investigate the effect of EID in more detail, Supplementary Figure S4 shows temporal slices of ΔR at the LEP (left) and HEP (right). In our computations, we have performed a switch-off analysis by including or neglecting EID effects. In each frame, the experimental (red-solid) and computed (black-solid: with EID effects; blue-dashed: without EID effects) spectra are compared. These slices are also indicated by black-solid lines along the HEP and LEP energies in Figs. 3a and 4a of the main manuscript and Supplementary Figure S3, respectively. While the full computation including EID effects (black-solid line in Supplementary Figure S4), shows excellent agreement with the experimental observations (red-solid), a computation without EID effects (blue-dashed) shows significant deviations from our measurements; besides changes of the overall amplitude, we observe much more pronounced oscillations if EID is not taken into account. In particular, the HEP experiences a stronger damping than the LEP because the higher-lying exciton states exhibit direct influence mainly on the HEP. Based on these comparisons, we conclude that our sample clearly exhibits excitation-induced shifts of the exciton-energy levels. Besides this, we observe that the oscillation period 0.66 ps (shaded area) is reproduced in all cases and not influenced by state-dependent broadening and shifting. This confirms that EID effects do not compromise our pump-reset protocol.

4. CAVITY DETUNING DEPENDENT PUMP-RESET

The pump-reset via THz interactions with the exciton-polaritons is susceptible to the cavity detuning δ_c . The exciton-polaritons are partly photons and partly excitons, where only the excitonic components are responsible for the THz-induced nonlinear optical effects because of no interaction between optical and THz photons. The relative portion of the excitonic component depends on the cavity detuning. The Hopfield coefficients $|X_L|^2$ and $|X_H|^2$ of the LEP and HEP modes quantify the relative exci-

Fig. S4. Experiment-theory comparison of differential reflectivity. Slices through low-energy peak (LEP, left) and high-energy peak (HEP, right) of the experimental (red-solid) and theoretical (black: with EID; blue-dashed: without EID) differential reflectivity $\Delta R = R_0 - R_{\text{reset}}$ as a function of delay between optical pump and THz-reset pulse. The slices shown here are also indicated by the black lines on LEP and HEP of Figs. 3a and 4a in the main manuscript, respectively, as well as Supplementary Figure S3. The oscillation period is 0.66 ps (shaded area), corresponding to the LEP-HEP energy splitting of 6.3 meV.

tonic content as a function of the cavity detuning [21, 22]:

$$|X_L|^2 = 1 - |X_H|^2 = \frac{1}{2} \left(1 + \frac{\delta_c}{\sqrt{\delta_c^2 + 4\hbar^2\Omega_0^2}} \right), \quad (4)$$

where $\hbar\Omega_0 = 3.15$ meV. Figure S5 shows (a) the experimental results and (b) the theoretical calculations of the THz-induced spectral modulations of the LEP and HEP modes with $E_{\text{THz}} = 10$ kV/cm at $\Delta t = 1.0$ ps for $\delta_c = -1.4, -0.9, -0.1, 0.4$, and 1.6 meV, which correspond to $|X_L|^2 = 0.39, 0.43, 0.49, 0.53$, and 0.62 , respectively. The nonlinear effects on the LEP mode enhance as δ_c increases, mainly because of the growing exciton proportion. Within the detuning range, the LEP mode is largely affected by nonresonant nonlinear effects, while resonant excitations from HEP to $2p$ -state dominate the HEP spectral modulations driving nearly complete mode bleaching regardless of δ_c .

REFERENCES

1. J. L. Tomaino, A. D. Jameson, Y.-S. Lee, G. Khitrova, H. M. Gibbs, A. C. Klettke, M. Kira, and S. W. Koch, "Terahertz Excitation of a Coherent Λ -Type Three-Level System of Exciton-Polariton Modes in a Quantum-Well Microcavity," *Phys. Rev. Lett.* **108**, 267402 (2012).

Fig. S5. Cavity detuning dependent THz-reset. (a) Experimental results and (b) theoretical calculations for THz-reset of the exciton-polariton modes for $\delta_c = -1.4, -0.9, -0.1, 0.4$ and 1.6 meV. The peak THz field amplitude is 10 kV/cm.

2. M. Kira, F. Jahnke, W. Hoyer, and S. W. Koch, "Quantum theory of spontaneous emission and coherent effects in semiconductor microstructures," *Prog. Quantum Electron.* **23**, 189–279 (1999).
3. G. Khitrova, H. M. Gibbs, F. Jahnke, M. Kira, and S. W. Koch, "Nonlinear optics of normal-mode-coupling semiconductor microcavities," *Rev. Mod. Phys.* **71**, 1591–1639 (1999).
4. H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 2009), 5th ed.
5. J. T. Steiner, M. Kira, and S. W. Koch, "Optical nonlinearities and Rabi flopping of an exciton population in a semiconductor interacting with strong terahertz fields," *Phys. Rev. B* **77**, 165308 (2008).
6. B. Ewers, N. S. Köster, R. Woscholski, M. Koch, S. Chatterjee, G. Khitrova, H. M. Gibbs, A. C. Klettke, M. Kira, and S. W. Koch, "Ionization of coherent excitons by strong terahertz fields," *Phys. Rev. B* **85**, 075307 (2012).
7. M. Kira and S. W. Koch, *Semiconductor Quantum Optics* (Cambridge Univ. Press, Cambridge (England, UK), 2011), 1st ed.
8. J. R. Danielson, Y.-S. Lee, J. P. Prineas, J. T. Steiner, M. Kira, and S. W. Koch, "Interaction of Strong Single-Cycle Terahertz Pulses with Semiconductor Quantum Wells," *Phys. Rev. Lett.* **99**, 237401 (2007).
9. M. Kira, W. Hoyer, and S. W. Koch, "Microscopic theory of the semiconductor terahertz response," *Phys. Status Solidi B* **238**, 443–450 (2003).
10. M. Kira and S. W. Koch, "Many-body correlations and excitonic effects in semiconductor spectroscopy," *Prog. Quan-*

- tum Electron. **30**, 155–296 (2006).
11. M. Teich, M. Wagner, D. Stehr, H. Schneider, M. Helm, C. N. Böttge, A. C. Klettke, S. Chatterjee, M. Kira, S. W. Koch, G. Khitrova, and H. M. Gibbs, “Systematic investigation of terahertz-induced excitonic Rabi splitting,” *Phys. Rev. B* (2014). accepted for publication.
 12. J. C. Maxwell, “A dynamical theory of the electromagnetic field,” *Philos. Trans. R. Soc. Lond.* **155** (1865).
 13. F. Jahnke, M. Ruopp, M. Kira, and S. W. Koch, “Ultrashort pulse propagation and excitonic nonlinearities in semiconductor microcavities,” in “Advances in Solid State Physics 37,” , vol. 37 of *Advances in Solid State Physics*, R. Helbig, ed. (Springer), pp. 191–206.
 14. H. Wang, K. Ferrio, D. G. Steel, Y. Z. Hu, R. Binder, and S. W. Koch, “Transient nonlinear optical response from excitation induced dephasing in GaAs,” *Phys. Rev. Lett.* **71**, 1261–1264 (1993).
 15. T. Rappen, U.-G. Peter, M. Wegener, and W. Schäfer, “Polarization dependence of dephasing processes: A probe for many-body effects,” *Phys. Rev. B* **49**, 10774–10777 (1994).
 16. P. Borri, W. Langbein, J. M. Hvam, and F. Martelli, “Well-width dependence of exciton-phonon scattering in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ single quantum wells,” *Phys. Rev. B* **59**, 2215–2222 (1999).
 17. B. Mieck, H. Haug, W. A. Hügel, M. F. Heinrich, and M. Wegener, “Quantum-kinetic dephasing in resonantly excited semiconductor quantum wells,” *Phys. Rev. B* **62**, 2686–2695 (2000).
 18. W. Schäfer, R. Löwenich, N. A. Fromer, and D. S. Chemla, “From Coherently Excited Highly Correlated States to Incoherent Relaxation Processes in Semiconductors,” *Phys. Rev. Lett.* **86**, 344–347 (2001).
 19. M. Kira and S. W. Koch, “Many-body correlations and excitonic effects in semiconductor spectroscopy,” *Prog. Quantum Electron.* **30**, 155–296 (2006).
 20. R. P. Smith, J. K. Wahlstrand, A. C. Funk, R. P. Mirin, S. T. Cundiff, J. T. Steiner, M. Schafer, M. Kira, and S. W. Koch, “Extraction of Many-Body Configurations from Nonlinear Absorption in Semiconductor Quantum Wells,” *Phys. Rev. Lett.* **104**, 247401 (2010).
 21. J. J. Hopfield, “Theory of the Contribution of Excitons to the Complex Dielectric Constant of Crystals,” *Physical Review* **112**, 1555–1567 (1958).
 22. H. Deng, H. Haug, and Y. Yamamoto, “Exciton–polariton Bose–Einstein condensation,” *Rev. Mod. Phys.* **82**, 1489–1537 (2010).