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Doubly dressed bosons – exciton-polaritons in a strong terahertz field

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We demonstrate the existence of a novel quasiparticle: an exciton in a semiconductor doubly dressed with two photons of different wavelengths: near infrared cavity photon and terahertz (THz) photon, with the THz coupling strength approaching the ultra-strong coupling regime. This quasiparticle is composed of three different bosons, being a mixture of a matter-light quasiparticle. Our observations are confirmed by a detailed theoretical analysis, treating quantum mechanically all three bosonic fields. The doubly dressed quasiparticles retain the bosonic nature of their constituents, but their internal quantum structure strongly depends on the intensity of the applied terahertz field.

The research on light – matter interaction in the strong coupling regime, when quantum light emitter and photons can coherently exchange energy, before the coherence is lost, is one of the fundamental problems in cavity quantum electrodynamics (QED). This problem was widely adopted in an atom – cavity system, described within renowned Jaynes-Cummings model and even beyond this limit, in the ultra-strong coupling regime. The system based on exciton-polaritons, quasiparticles composed from excitons in a semiconductor strongly coupled to the vacuum light field in the cavity, has many advantages. These quantum light emitters do not show fermion-like statistics, but are truly designed bosons that can exhibit non-equilibrium Bose-Einstein phase transition [1]. In the present manuscript, we study the phenomenon of double dressing: an exciton coupled to two photonic fields from distinct energy ranges: near-infrared (NIR) and terahertz (THz), which bares no direct analogy in atomic physics.

Excitons, or bound electron-hole pairs, can be created by the absorption of a near infrared (NIR) photon in a semiconductor. The strong coupling [2–4] of excitons and photons in a high-quality microcavity structure results in the formation of exciton-polaritons, due to vacuum field Rabi coupling, evidenced by the appearance of lower polariton (LP) and upper polariton (UP) resonances [5]. The strong coupling regime can be achieved when the energy exchange rate between the states is larger than the decoherence rate. Then the description of the system can be properly made in terms of new quantum eigenstates, or dressed quasiparticles. In a quantum well (QW), which confines the exciton into a plane, the internal structure of the exciton resembles that of a two-dimensional hydrogen atom. The transitions between the internal states lie in the THz range of the electromagnetic spectrum. The possibility to induce such transitions with THz photons was shown in Refs. 6–8. Upon intense THz illumination, close to the $1s$-$2p$ excitonic transition, Autler-Townes splitting of excitonic states has been observed [9, 10] together with the possibility to imprint the coherent phase of the driving field [11]. Also the strong influence of an exciton-polariton reservoir was revealed upon THz excitation of an exciton-polariton condensate [12].

In this letter we demonstrate the simultaneous dressing of excitons with NIR and THz photons. We observe the appearance of a third dressed polariton mode, the middle polariton (MP), which is accompanied by an energy shift of the upper and lower polariton states. We describe our observations with a quantum model that takes into account the relevant couplings between four bosonic fields, including $1s$ and $2p$ excitons together with NIR and THz photons. Previously, doubly dressed states of a two-level quantum dot system with two optical photons [13] were shown, as well as interaction of exciton-polaritons with THz photons [14]. However, quantum dots exhibit fermionic-like single-photon emission and photon anti-bunching. By contrast, exciton-polaritons are bosonic particles, which also applies to our new quasi-particle observed here.

To confine the NIR photonic mode we used a GaAs lambda microcavity sandwiched between two AlAs/GaAs distributed Bragg reflectors. A single 8 nm-thick $In_{0.04}Ga_{0.96}As$ quantum well was placed, at the maximum of the cavity field, providing the excitonic component of polaritons. Excitons confined inside the quantum well couple to the photon modes with a coupling strength given by the Rabi splitting, Ω_C. In the case of our sample, the excitonic resonance was at approximately 1.484 eV and the vacuum Rabi splitting was equal to approximately $\Omega_C = 3.5$ meV. The on-resonance polariton linewidth was 0.3 meV allowing for a clear resolution of polariton states. Details on the sample structure and optical characterization can be found in Ref. 15 and

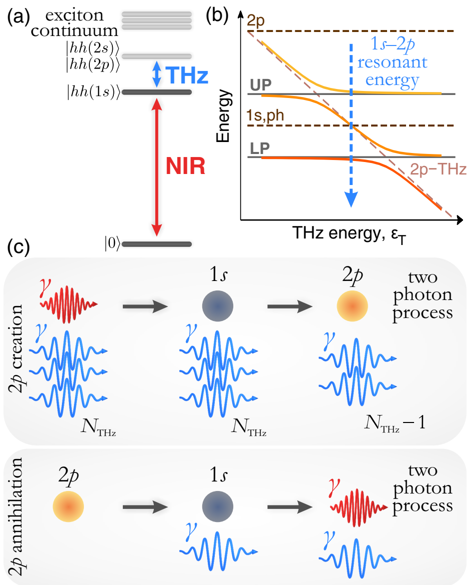

FIG. 1: a) Scheme of the internal structure of an atom-like exciton. The vacuum state ($|0\rangle$) is coupled to a ground $1s$ heavy hole (hh) exciton via the NIR photon. The $1s$ is coupled to the excited $2p$ exciton via THz photon. In our theoretical model we neglect other excited electronic states, but we include the exciton ionization and blueshift from the dynamical Franz-Keldysh effect. b) The bare exciton-polariton energies (black solid lines) and the double dressed quasiparticle energy diagram (yellow-orange lines). The THz energy resonant to the $1s$–$2p$ transition is marked with a blue arrow. c) Scheme of the creation and annihilation of the $2p$ exciton state. This process involves two photons, one from NIR and one from THz spectral ranges.

Ref. 16, respectively. The sample was kept in a cryostat at the temperature of 6 K. The polariton population was created resonantly by fs laser pulses, with the spot size of 150 μm. The pulse spectrum was sufficiently broad to simultaneously excite LP and UP branches at zero momentum. In the experiments reported here, the excitation power was low enough, approx. 1.5 μW average power, to maintain polaritons in the linear regime of polariton-polariton interactions. All experimental observations are reported for close to zero exciton-cavity photon detuning $\delta \in (-0.2, 0.2)$ meV.

The THz light was generated at the Helmholtz-Zentrum Dresden-Rossendorf by the free-electron laser (FEL). The spectral width of the pulse at the resonant wavelength of 182 μm was 1.6 μm (which corresponds to (6.80 ± 0.03) meV). The THz pulse duration was determined to be approx. 30 ps [10], and the spot size was about 1 mm in diameter. Since this is significantly larger than the NIR spot size, the THz power can be regarded to be uniform inside the probed area. The scheme of the experimental setup is provided in the Supplementary Material (SI).

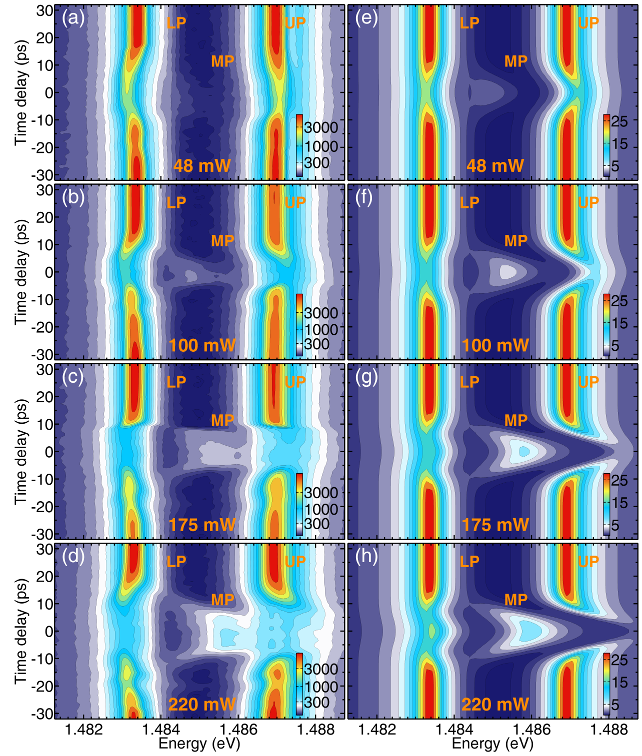

FIG. 2: Transmission spectra of lower (LP) and upper (UP) polaritons (at normal incidence) as a function of the delay between the NIR and THz pulses (in equidistant time intervals). The color scale indicates the intensity in arbitrary units. The THz photon energy is 6.8 meV, slightly detuned from $1s$–$2p$ excitonic transition (6.5 ± 0.5 meV). The appearance of THz induced middle branch (MP) at the maximum of THz pulse (zero delay) is visible. This is accompanied by an energy shift of LP and UP branches. a)- d) Illustrates the behavior with increasing average THz power as marked directly in the image. Positive delays correspond to the NIR pulse preceding the THz pulse, negative delays to the reversed situation. e) - h) Corresponding theoretical model. The color scale is nonlinear to enhance the low-intensity signal at zero delay time and the background signal was removed from the figures.

The exact energy separation between the $1s$ and $2s$ exciton states was determined experimentally [17]. We have observed the appearance of the $2s$ exciton-polariton in photoluminescence spectra in magnetic fields. From the observed energy change in magnetic fields we could determine the zero field energy splitting between $1s$ and $2s$ exciton in our sample to approx. 6.5 meV. Moreover, at zero magnetic field $2s$ and $2p$ states are very close in energy and we can estimate the energy difference between $1s$ and $2p$ state to 6.5 ± 0.5 meV. Our measurement results agree with other works performed on similar structures [10].

The idea of the experiment is as follows: a weak NIR laser pulse creates a population of exciton polaritons through the excitation of an initially empty semiconductor QW inside a microcavity (Fig. 1a). A strong THz

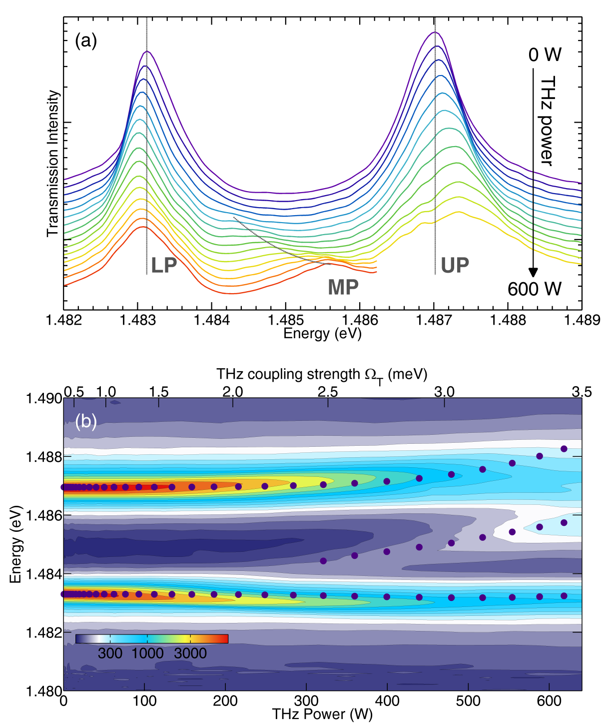

FIG. 3: Transmission spectra (at normal incidence) of semiconductor microcavity revealing dressed polaritons and approaching to ultra-strong coupling regime. a) Spectra illustrate the cross section of the results shown in Fig. 2d for the time delays from 15 ps to 0 ps. At weak perturbation (at 15 ps time delay, that corresponds to the onset of THz pulse) the transmission spectra reveals unperturbed LP and UP states. The time delay was converted to instantaneous THz power, as indicated directly in the image, based on Fig. 3 from SI. As THz power increases, a red-shift of the LP and a blue-shift of the UP are observed and the additional MP line is visible (the dotted lines are guides to eye to compare the energy shifts, for vertical lines the energy is constant, the middle dotted line follows the MP energy shift). The THz induced opacity of the sample is revealed by the decrease of the overall intensity of the transmitted signal. b) Comparison of the experimental data from Fig. 2d (colored map) with theoretical model from Fig. 2h (dots). The interaction energy with the THz field is illustrated in the top scale.

external laser pulse, tuned to resonance with the $1s$ to $2p$ transition, mixes the two excitonic states, as shown in Fig. 1b. Fig. 1c illustrates the $2p$ excitation and decay scheme. The two external laser beams are collinear and synchronized. The NIR Ti:Sapphire laser provides 120 fs pulses, with central wavelength tuned to the excitonic resonance. The spectral width was broad enough to cover both LP and UP. Therefore exciton-polaritons were created resonantly in a superposition of polariton states.

The NIR transmission spectra taken for normal incidence as a function of the delay time between the NIR and THz pulses are reported in Fig. 2. When the system is unperturbed by the THz beam, i.e. at large positive or negative delays, we observe the two expected transmission resonances corresponding to LP and UP polaritons as in Ref. 16 and in Supplemental Information (SI). For delays of the order of 15 ps, the THz pulse intensity starts to increase and the energy and the intensity of both polariton lines are modified. The energy of LP and UP are repelled apart, which is visible already at low average THz power, Fig. 2(a,e). At slightly higher average THz power, Fig. 2(b,f), the LP and UP energy shift is faster and one additional line appears between polariton states, getting stronger for larger THz powers, Fig. 2(c,g) and (d,h). We will refer to this line as the middle polariton branch (MP).

In all cases, the LP and UP energy shifts and the appearance of the MP are accompanied by a drastic decrease in the signal intensity, especially pronounced for exact time overlap between the pulses where the sample is getting less transparent. A THz average power as high as 200 mW gives an electric field strength of 12 kV$\cdot$ cm^{-1} (or intensity of 1 MW$\cdot$ cm^{-2}) at the position of the QW. In such a regime of THz excitation the excitons can ionize.

The appearance of the middle polariton line should in no case be considered as an indication of the transition to the weak coupling regime [18] and the saturation of the exciton transition. This is confirmed by the following: a) all three lines coexist at a given time and THz power; b) simultaneous energy shift of LP, UP and MP with increasing THz power (the energy of cavity photon is independent on the THz power); c) the MP energy depends on the THz power (Fig. 3a) and THz detuning from $1s$-$2p$ transition (see SI). All images illustrated in Fig. 2 are symmetric versus zero delay time, which demonstrates that even at very high THz powers the exciton population is not destroyed. Indeed, we are in the low NIR excitation regime and the total excited exciton population is far from the saturation density.

To understand the physics behind our observations, we performed calculations based on the four-mode Hamiltonian. The derivation (described in the SI) is based on the rotating wave approximation, focusing on the four resonantly interacting bosonic modes. A $1s$ exciton can be formed by absorption of a NIR photon. The $1s$ exciton can further absorb a photon from the external THz field to form an excited $2p$ state, see Fig. 1c. The annihilation process of a $2p$ exciton is a reverse two photon process. These are described by

$$H = \sum_{\nu=1s,2p,C,T} \epsilon_\nu \hat{a}_\nu^\dagger \hat{a}_\nu + \hbar\Big(g_C \hat{a}_C \hat{a}_{1s}^\dagger + g_T \hat{a}_T \hat{a}_{1s} \hat{a}_{2p}^\dagger + + g_C \hat{a}_C^\dagger \hat{a}_{1s} + g_T \hat{a}_T^\dagger \hat{a}_{1s}^\dagger \hat{a}_{2p} \Big) + H_{\mathrm{high}}. \quad (1)$$

Here, $\hat{a}_{1s}, \hat{a}_{2p}, \hat{a}_C$, and $\hat{a}_T$ are bosonic annihilation operators for the $1s$ and $2p$ excitons, cavity and THz photons, and $\epsilon_{1s,2p,C,T}$ are their respective energies. The coupling of the $1s$ excitons to photons is given by g_C, while g_T determines the probability of the $1s$–$2p$ transition with simultaneous annihilation of a THz photon. The $H_{\rm high}$ term describes coupling to other electronic excitations.

This Hamiltonian does not provide a simple bosonic quasiparticle spectrum, even when neglecting this last term. However, as shown in the SI, when the occupation of the THz mode is large, the elementary bosonic excitations (dressed states) turn out to be the solutions of

$$H_{single} = \begin{pmatrix} \epsilon_C & \Omega_C^*/2 & 0 \\ \Omega_C/2 & \epsilon_{1s} & \Omega_T^*/2 \\ 0 & \Omega_T/2 & \epsilon_{2p} - \epsilon_T \end{pmatrix} \qquad (2)$$

where $\Omega_C = 2\hbar g_C$, $\Omega_T = 2\hbar g_T\sqrt{N_T}$, with N_T the average occupation of the THz laser mode, $\Delta = \epsilon_{2p} - \epsilon_{1s} - \epsilon_T$ is the detuning of the THz field, and $\delta = \epsilon_C - \epsilon_{1s}$ is the exciton-photon detuning. Note that the $1s$ exciton-cavity coupling Ω_C is independent of the number of photons, and corresponds to the vacuum Rabi splitting [19]. In contrast, the $1s$–$2p$ coupling Ω_T is proportional to the square root of the number of photons in the coupling THz field, which is in analogy with the Autler-Townes effect [20]. The above matrix has three eigenstates, which correspond to lower, middle, and upper polaritons, as illustrated in Fig. 1d. In the absence of a strong THz field, one of the modes corresponds to the excitation of the uncoupled, optically inactive bare $2p$ exciton. Under intense THz field, this mode becomes dressed with optically active states and becomes increasingly visible as the MP spectral line in Fig. 2. We note, however, that all three lines correspond to doubly dressed states, since all three are composed of exciton, photon and THz excitations.

We reproduce the experimental spectra by computing the solutions of (2), while including pumping and losses (see SI). The effects coming from the higher electronic states $H_{\rm high}$ can be included, with a good accuracy, when considering two phenomena. The effect of exciton ionization [21, 22] by the strong electric field of the THz laser leads to the decay of the LP and UP lines at high values of the THz power, as visible in Fig. 2. At the same time, the dynamical Franz-Keldysh effect increases the energy of the exciton states due to the induced motion of electron and hole, which is most clearly seen in the asymmetry of the LP and UP energy shifts (Fig. 3).

Fig. 3a illustrates a few NIR transmission spectra that correspond to the cross sections of the map from Fig. 2d. Fig. 3b shows the exact comparison of the experimental data (Fig. 2d) with the theoretical model (Fig. 2h). The time-averaged THz pulse power as high as 200 mW corresponds to $N_T = 1.4 \times 10^{13}$ photons in a single coherent pulse. As the THz coupling strength, $\Omega_T \propto \sqrt{N_T}$, the dipole interaction energy (top scale in Fig. 3b) can approach the same order as the transition energy (approx. 6.5 meV). Therefore, we can tune the strength of the interactions through all regimes, from weak to ultra strong [23].

In conclusion, strong coupling involving both the vacuum field Rabi splitting and the coupling to a second propagating field gives rise to a novel class of coherent phenomena in the solid-state, where the open dissipative character of the system competes with strong coherence. Bosons doubly dressed with optical and THz fields are of fundamental interest due to the nontrivial internal structure of the quantum state. Our states are superpositions of NIR photons, THz photons and excitons. We have shown here that the dressed particle description remains valid, albeit with the addition of a third dressed state in the picture. The coupling to a strong coherent THz field might be considered in the design of semiconductor-based THz devices [24, 25]. Possible blocking of the NIR transmission of the micro-cavity structure induced by the THz beam allows one to consider applications such as ultrafast optical switches. Condensation of doubly dressed polaritons in a THz field may be interesting due to the possibility of achieving dipole-dipole interactions in condensates with admixture of $2p$ states. This may lead to interesting new phenomena such as supersolidity [26].

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