Intersubband polaritonics

The interaction of light with material excitations has long attracted intense experimental and theoretical investigations. Thanks to the development of epitaxial growth techniques, it is now possible to realize monolithic microcavity structures in which both electrons and photons can be simultaneously confined and their interaction can be manipulated. In a microcavity, the photonic confinement deeply modifies light-matter interaction, and if the coupling strength is sufficiently large compared to the damping rates, new elementary excitations are formed which are mixed electronic and photonic states. The so-called "cavity polaritons" were first observed in solid-state systems employing excitonic states in semiconductors. Recently we demonstrated the strong coupling of the confined electromagnetic radiation with the transitions between the subbands in the conduction band of a GaAs-AlGaAs heterostructure, with the corresponding formation of "intersubband cavity polaritons" [1]. This system is very promising for the achievement of an unprecedented "ultra-strong coupling regime" of light-matter interaction, which is particularly interesting for the peculiar quantum nature of its eigenstates. Furthermore, theoretical predictions show that the ultrafast modulation of polariton coupling can result in the release of correlated photon pairs from the polariton ground state [2], in a process analogous to the dynamic Casimir effect and to Hawking black-hole radiation. Here we present evidence of cavity polaritons established between excited subbands and discuss the measurement of their dispersion [3]. First signatures of ultra-strong coupling contributions to the polariton energies are also reported [4] and non-adiabatic sub-cycle modulation of the coupling strength is demonstrated [5].

In an intersubband microcavity, the vacuum Rabi energy is a measure of the electromagnetic coupling and determines the polaritons energy splitting. It scales as the ratio $\langle d_{ISBT} \rangle / \sqrt{L_{QW}}$ between the dipole matrix element $\langle d_{ISBT} \rangle$ and the quantum well thickness L_{QW} . By judicious design, in larger quantum wells, one can have an increased using the transition from the first excited subband to the next higher subband (levels 2 and 3 in the inset of Fig.1), if the wells are doped so that the electron Fermi energy E_F is above level 2. The

angle-resolved reflectance measurements of a 70° prism-shaped sample with 3.7×10^{18} cm⁻³ n-doped active region at liquid helium temperature are reported in Fig. 1. Two dips corresponding to the lower and upper polaritons are clearly observed, exhibiting a minimum splitting of about 40 meV, at an internal angle of about 63.5°. The splitting increases to about 52 meV at 300 K [3]. In order to extract precise information on the strength of the optoelectronic interaction, one should express the polariton dispersion



Fig. 1 Left: reflectance spectra of the microcavity sample resonant with the excited subband transition (2-3) as a function of incidence angle. Right: energy-wavevector dispersion; the dashed lines correspond to the uncoupled cavity and transition energies; experimental points are plotted as open circles; the shaded area outside the light cone is not accessible with the

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Fig. 2

Angle-resolved reflectance of the intersubband microcavity displayed as contour plot (arb. units). The superimposed curves shown as circles are theoretical fits with the Rabi energy as the only free parameter. White curves were calculated using the full Hamiltonian, red ones without the anti-resonant term, and black ones with neither photon energy renormalization nor anti-resonant contributions. The right panel reports the reflectance spectrum corresponding to the internal angle of 68.5°. Arrows show the calculated lower polariton energies for the three Hamiltonians detailed above



as function of the in-plane wavevector $k_{//}$. The necessity arises from the fact that the polariton peaks at the same internal angle do not correspond to the same $k_{//}$. The actual energy-wavevector dispersion is reported in Fig. 1. As the lower polariton curve cannot be measured up to the anti-crossing point, an accurate fitting procedure is necessary in order to obtain the value of the vacuum-Rabi energy. The solid lines in Fig. 1 show the results of fitting the experimental data with the complete quantum-mechanical theory detailed in Ref. 2. The same procedure has also been performed on a different sample, operating though conventionally between the first two subbands. The measured reflectance is compared in Fig. 2 with the computed curves of the polariton dispersion [4]. Experimental data are shown as contour plot, while the polariton energies calculated from the full Hamiltonian are shown as white circles. A perfect agreement is found for a vacuum Rabi energy $\hbar\Omega = 17$ meV, i.e., ~ 11 % of the intersubband transition energy (152 meV). Black circles in Fig. 2 were instead obtained using the same value of Ω but neglecting both diamagnetic and antiresonant terms (defining the ultra-strong coupling), while the red ones excluding only the anti-resonant contribution but

including the photon renormalization. Though only small deviations appear for the upper polariton, the lower polariton dispersion calculated with either of the latter approximations is clearly incompatible with the experimental measurements, proving the relevance of these anomalous contributions [4].

As a consequence of the anti-resonant terms, the ground state of the system is a squeezed vacuum containing a finite number of virtual photons. Theory shows that non-adiabatic switching of Ω_R may release these virtual quanta in correlated pairs [2]. As a matter of fact, non-adiabatic control poses an extreme experimental challenge: changes are required to be induced on a scale set by the cycle of light. We introduced then an all-optical scheme for femtosecond control of ultrastrongly coupled cavity polaritons - a first practical test-bed to target non-adiabatic QED phenomena [5]. Our sample contains undoped GaAs QWs, and radiative transitions between the subbands only become possible if electrons are injected extrinsically. We employ a 12-fs control pulse to excite electrons resonantly from the valence band into the first conduction subband, activating the intersubband field within femtoseconds. The eigenmodes of this system are then probed by resonant



reflection of a second mid-IR light pulse. Fig. 3a displays amplitude spectra recorded at various delay times t_D. The initial bare photon state is replaced by two coupled polariton branches appearing simultaneously at energy positions of 93 meV and 143 meV. Most remarkably, the new resonances do not gradually develop out of the bare cavity mode, but the cavity-intersubband coupling forms quasi-instantaneously.

The physics is most striking in the time domain. When a few-cycle multi-THz transient (time trace in Fig. 3b) impinges on the unexcited modulator structure, part of its energy is directly reflected off the cavity surface. A second portion is transmitted into the structure, prepares a coherent photon state in the resonator, and gets re-emitted subsequently contributing to the overall reflected signal. This dynamics is encoded in a characteristic twin-pulse temporal structure of the reflected transient (Fig. 3c). The most intriguing situation is realized if we switch the eigenstates of the cavity while a coherent state of bare photons is still present (Fig. 3d): the incident THz transient first prepares a photon population in the unexcited cavity. During the free emission decay, the control pulse abruptly alters the eigenstates of the system. Remarkably, the decay of the bare cavity mode is interrupted on a time scale shorter than the oscillation cycle of light - a compelling proof of non-adiabaticity. The subsequent reemitted transient exhibits a characteristic two-mode beating in the time domain. The corresponding spectrum displays minima at photon energies of 93 meV and 143 meV - the hallmarks of both polariton branches.

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