Polaritonic Cross Feshbach Resonance

M. Navadeh-Toupchi,* N. Takemura, M. D. Anderson, D. Y. Oberli, and M. T. Portella-Oberli
Institute of Physics, School of Basic Sciences, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

(Received 13 June 2018; revised manuscript received 22 October 2018; published 31 January 2019)

We demonstrate the existence of a cross Feshbach resonance by strongly driving a lower polariton mode and by monitoring in time the transmission of a short optical pulse at the energy of the upper polariton mode in a semiconductor microcavity. From the signatures of the optical resonance, strength, and sign of the energy shift, we attribute the origin of the scattering process between polariton modes with opposite circular polarization to a biexciton bound state. From this study, we infer the conditions required for a strong enhancement of the generation of entangled photon pairs.

DOI: 10.1103/PhysRevLett.122.047402

A Feshbach resonance appears whenever two free particles resonantly couple to a molecular bound state. Near the resonance, the strength of the interaction between the particles is modified, and its sign changes at resonance. Since the demonstration of atomic Feshbach resonances [1,2], they have been extensively used to control the interactions in atomic Bose Einstein condensates, and they have been the key to many breakthroughs [3–7]. Recently, the Feshbach resonance has been implemented to tune the interaction strength between a mobile impurity and a Bose gas of cold atoms in order to realize a Bose polaron in a strongly interacting regime [8,9]. The fascinating control of the interaction strength may also be implemented directly with a system of polaritons in a semiconductor microcavity. A polaritonic Feshbach resonance was demonstrated in a semiconductor system when two lower polaritons were efficiently coupled to the biexciton, which is the quasiparticle analogue of the molecular state in a semiconductor [10]. The polariton-polariton interaction strength was tuned by involving only polaritons from the lower polariton branch. Likewise, one would expect a similar enhancement of the scattering strength when polaritons from both the upper and lower branches are involved through a polaritonic cross Feshbach resonance. This demonstration will permit the control of the polariton interbranch scattering, and it may initiate studies of many-body physics with polaron quasiparticles and lead to the generation of entangled photon pairs via the biexciton. In the former case, this system may share similar polaron properties of an atomic impurity interacting with a Bose gas, where the upper polariton mode replaces the impurity atom and the coherent population of lower polaritons substitutes for the Bose gas. In the latter case, different schemes for generating pairs of entangled photons in semiconductor have been the subject of several theoretical studies [11–15]; for the most effective one, it relies on the generation of biexcitons by interbranch polariton scattering. Despite the great interest attracted by sources of non-classical light, for quantum cryptography [15] and quantum information processing [16], an implementation via the biexciton in an optical microcavity is still pending. It will hold its promise, however, if an efficient generation of biexcitons could be demonstrated by enhancing the interbranch scattering of polariton pairs.

In a semiconductor microcavity, the strong coupling between excitons and photons gives rise to two new eigenstates: the lower (LP) and the upper polaritons (UP) [17]. These quasiparticles provide nonlinear behavior due to excitonic interactions and coherent properties because of their photonic component, which has made them attractive to study phenomena such as polariton condensates [18,19], quantum fluids [20,21], quantized vortices [22], Dirac cones [23], spontaneous spin bifurcations [24], and squeezing [25,26].

The spinor character of polariton interactions [27–29] offers a wide range of physics to explore [30–32]. For instance, two excitons with opposite spins can form a molecular bound state, which is called a biexciton. Polaritonic Feshbach resonance [10] allows the modification of lower-polariton self-interactions from attractive to repulsive when tuning the energy of the two polaritons across the resonance with the biexciton.

The exciton-exciton interaction not only introduces polariton self-interactions (LP-LP) but also cross polariton interactions (LP-UP) [33,34]. It was theoretically predicted [13,14] that a biexciton is generated by the scattering of one lower and one upper polariton and decays into two lower

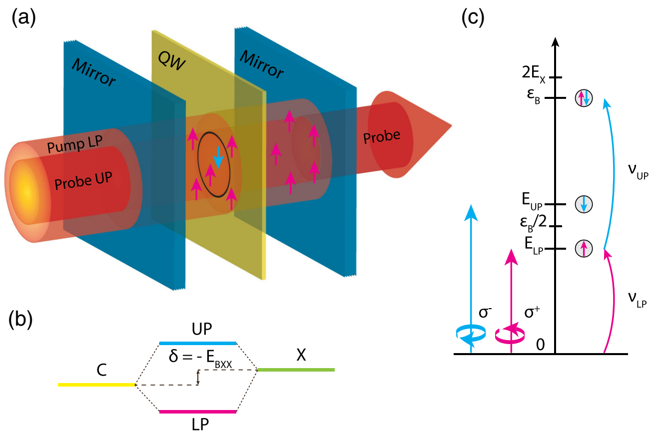

FIG. 1. (a) A schematic view of the experimental system comprising a semiconductor microcavity with a quantum well in its center. (b) The cross Feshbach resonance occurs at the cavity detuning between the exciton state (X) and the cavity mode (C) given by $\delta = -E_{\mathrm{BXX}}$, E_{BXX} is the biexciton binding energy. (c) Under this condition, an upper polariton with a spin down ($\downarrow$) and a lower polariton with a spin up ($\uparrow$) scatter resonantly to the biexciton state ($\downarrow\uparrow$). E_X, ε_B, E_{UP} and E_{LP} are, respectively, exciton, biexciton, upper-polariton, and lower-polariton energy. ν_{LP} (ν_{UP}) and σ^+ (σ^-) are, respectively, the energy and polarization of the pump (probe) pulse.

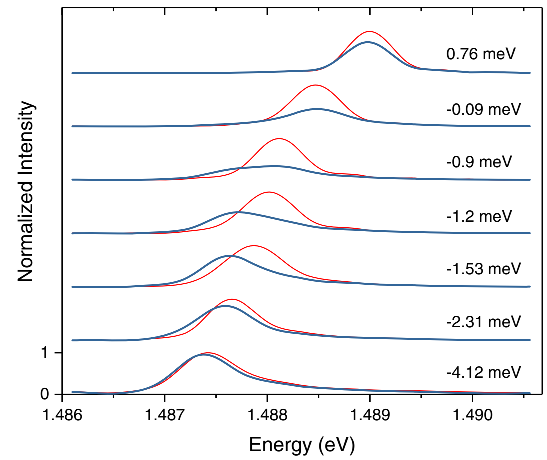

FIG. 2. Transmitted probe spectra at upper polariton peak with (blue, thick line) and without (red, thin line) the presence of the antiparallel spin lower polariton population for different detunings at $\tau = 0$.

polaritons with opposite momenta and spins, which emit a photon pair entangled in polarization and momentum. This process is expected to be optimal at exciton-cavity detuning comparable to the biexciton binding energy. This corresponds to the condition for which the energy sum of the lower and upper polaritons with opposite spins equalizes the biexciton energy. Under this condition, a cross Feshbach resonance is expected to be observable. The determination of this cross Feshbach resonance is thus, the main step towards the generation of entangled photon pairs.

In this Letter, we demonstrate the cross Feshbach resonance of a pair of lower and upper polaritons coupling to a biexciton in a semiconductor microcavity. We show a clear resonance for which, in the presence of a spinor lower-polariton population, the energy of an upper-polariton with an antiparallel spin undergoes abrupt renormalization from an attractive to a repulsive interaction. Correspondingly, the maximum reduction of the upper polariton emission shows the resonant conversion of the upper and lower antiparallel spin polariton pair into a biexciton. The cross Feshbach resonance is scrutinized by tuning the cavity-exciton energy. We show the very fast dynamics of the scattering process in which the cross interaction between lower and upper polaritons couples resonantly with the biexciton state.

We use spectrally resolved circularly polarized pump-probe spectroscopy. The lower polaritons are excited resonantly with a circularly polarized (σ^+) narrow-band pump pulse generating a spinor lower polariton population. The cross interaction between the upper and lower polariton is probed with a counter-circular polarized (σ^-) probe pulse [Fig. 1(a)]. We spectrally probe the energy and intensity of the upper polariton peak by measuring the transmission spectrum of the probe pulse. The cross Feshbach resonance occurs at a negative cavity detuning energy equal to the binding energy of the biexciton [Fig. 1(b)]. At this detuning, the total energy of one lower plus one upper polariton matches the biexciton energy [Fig. 1(c)]. The dynamics around the cross Feshbach resonance are studied by performing a time-resolved experiment.

The sample under investigation is a III-V GaAs based microcavity [35]. A single 8 nm $In_{0.04}Ga_{0.96}As$ quantum well is sandwiched between a pair of GaAs/AlAs distributed Bragg reflectors. The Rabi splitting at zero cavity-exciton detuning is $\Omega_R = 3.45$ meV and the exciton energy is $E_X = 1.4868$ eV. We perform a pump and probe experiment in a close degenerate configuration ($k_{\mathrm{probe}} \approx k_{\mathrm{pump}} = 0$). The broadband few hundred femtosecond pulses are generated by a Ti:Sapphire laser with an 80 MHz repetition rate, and the pump pulse is spectrally narrowed (to 0.5 meV) by a pulse shaper. The pump spot size is larger than the probe to ensure the probing of a constant carrier density. The probe intensity is one tenth of the pump $n^{\mathrm{pu}} = 2.8 \times 10^{11}$ photon pulse^{-1} cm^{-2}. The energy of the pump pulse is adjusted to the energy of the lower polariton resonance. The probe spectrum is measured in transmission as a function of the detuning and also as a function of the delay τ between pump and probe pulses. $\tau > 0 (\tau < 0)$ means that the pump (probe) arrives before the probe (pump) pulse. The sample is kept at a temperature of 4 K.

The character of the polariton interaction is detected through the energy shift of the polariton resonance: a blueshift or a redshift meaning, respectively, a repulsive or an attractive interaction. In Fig. 2, we compare the transmitted probe spectrum of the upper polariton resonance for

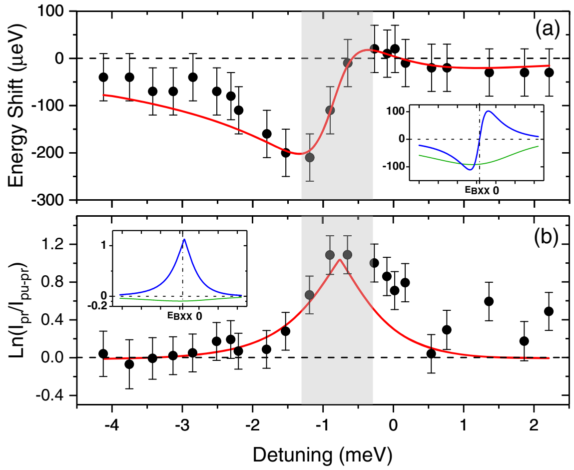

FIG. 3. Energy shift (a) and intensity variation (b) of the upper polariton peak in the presence of the antiparallel spin lower polariton population as function of cavity detuning. The dots and the solid lines are the experimental and numerical simulation results, respectively. The gray shaded area shows the detuning range where scattering to the biexciton is effective. Inserts: The two contributions of background (green, thin line) and biexciton scattering (blue, thick line) to the upper polariton energy shift (a) and absorbance (b) as a function of detuning.

different cavity detunings, measured at zero pump-probe delay, with or without the pump. We can track the changes in the energy shift of the upper polariton in the presence of an antiparallel spin lower polariton population. A change of detuning from negative to positive corresponds to an energy shift of the upper polariton being first a redshift, then switching to a blueshift and recovering a zero value; notice that the amplitude of the signal first decreases and then increases, and vice versa with the linewidth. In Fig. 3(a), we plot as a function of cavity detuning the energy shift and, in Fig. 3(b), the change of absorbance of the transmitted probe signal at zero time delay, $\ln(I_{\mathrm{probe}}/I_{\mathrm{pump-probe}})$, where I_{probe} and $I_{\mathrm{pump-probe}}$ refer to the maximum intensity of probe signal either alone or in the presence of the pump pulse. The signature of the cross Feshbach resonance is clearly evidenced at -0.9 meV detuning through the change of sign of the energy shift, which demonstrates the switching of the nature of the interactions between lower and upper antiparallel spin polaritons from attractive to repulsive. Correspondingly, the maximum reduction of the signal intensity at this detuning shows the resonant conversion of the upper and lower antiparallel spin polariton pair into a biexciton. This optimum cavity detuning also provides a direct measure of the biexciton binding energy E_{BXX} [see inserts in Figs. 3(a) and (b)]. Its value, $E_{\mathrm{BXX}} = 0.9$ meV, lies in the expected range within the three-dimensional limit (0.15 meV) and the two-dimensional limit (1.4 meV) given for an infinity deep confinement potential. The latter limit is estimated by using a ratio of 0.22 between the biexciton and the exciton binding energies [36] and an experimental value of the exciton binding energy of 6.5 meV [37]. The increased depletion rate of the upper polariton population is quantified by the absorbance [Fig. 3(b)]. The depletion rate caused by the upper-polariton-lower-polariton scattering to the biexciton at resonance corresponds to the relative change of the probe transmission at the energy of the upper polariton mode that is equal to 0.35 ($=1 - re^{-1.1}$), where 1.1 is the maximum of the measured absorbance change and $r = 2$ is the ratio of the upper polariton linewidths with and without the pump pulse. This means that about 35% of the transmitted photon flux of the probe pulse at the energy of the upper polariton gives rise to the creation of biexcitons at the cross Feshbach resonance.

On the basis of the theoretical model developed for lower polariton interactions in the vicinity of a Feshbach resonance described in [29], the detuning dependence of the probe beam transmission spectrum was obtained similarly by solving the coupled Gross-Pitaevskii equations of motion for upper and lower polariton modes and one Heisenberg equation of motion for the biexciton state. Assuming cw optical excitation, analytical expressions of the dependence with the cavity detuning can be obtained for the energy shift and for the transmitted probe amplitude of the upper polariton. The dependence of the upper polariton energy shift $\Delta E_{U,\downarrow}$ with the cavity detuning is given by:

$$\Delta E_{U,\downarrow} = g^{+-}X_0^2|C_0|^2|\psi_{L,\uparrow}^{\mathrm{pu}}|^2 + \mathrm{Re}\left(\frac{g_{\mathrm{bx}}^2 X_0^2|C_0|^2|\psi_{L,\uparrow}^{\mathrm{pu}}|^2}{\varepsilon_L + \varepsilon_U - \varepsilon_B + i\gamma_B}\right), \tag{1}$$

where X_0 and C_0 are the Hopfield coefficients and $\varepsilon_L + \varepsilon_U = 2E_X + \delta$. The antiparallel spin $\uparrow, \downarrow$ lower-upper polariton background interaction constant is given by $g^{+-}X_0^2|C_0|^2 = g_{UL}^{+-}$, and g_{bx} is the coupling strength to the biexciton. The lower polariton density generated by the pump is $|\psi_{L,\uparrow}^{\mathrm{pu}}|^2 = |C_0|^2 n^{\mathrm{pu}}$. ε_L and ε_U are the energy of the lower and upper polariton, ε_B and γ_B are the biexciton energy and linewidth. The binding energy of the biexciton is $E_{\mathrm{BXX}} = 2E_X - \varepsilon_B$.

There are also two contributions to the change of absorbance of the upper polariton mode in the presence of a population of lower polaritons excited by the pump pulse. The first one originates from the polariton-polariton scattering to the biexciton, and its dependence with cavity detuning is given by:

$$\alpha_B = g_{\mathrm{bx}}^2 X_0^2|C_0|^2|\psi_{L,\uparrow}^{\mathrm{pu}}|^2 \frac{\gamma_B}{(\varepsilon_L + \varepsilon_U - \varepsilon_B)^2 + \gamma_B^2}. \tag{2}$$

The second contribution results from the negative background term that corresponds to an attractive interaction between upper and lower polariton modes of opposite spins. This interaction results in an effective energy redshift of the exciton, $\Delta E_X = g^{+-} X_0^2 n^{\mathrm{pu}}$, and then in an effective increase of the cavity detuning $\delta' = \delta - \Delta E_X$, which implies a larger photonic fraction of the upper polariton mode [38]. The dependence of this change of absorbance with detuning is thus given by: $\alpha_b = 2\ln[X_0(\delta)/X_0(\delta')]$, where $X_0^2 = \frac{1}{2} + [\delta/(2\sqrt{\delta^2 + \Omega_R^2})]$.

In Figs. 3(a) and 3(b), we compare the detuning dependence of the experimental results with the dependence given by above expressions for the energy shift and for the absorbance, respectively. The best fit with the theoretical model was obtained with $E_{\mathrm{BXX}} = 0.9$ meV, $g_{\mathrm{bx}} = 0.86\ \mathrm{meV}/\sqrt{n^{\mathrm{pu}}}$, $\gamma_B = 0.5$ meV and $g^{+-} = -0.62\,\mathrm{meV}/n^{\mathrm{pu}}$. In the inserts of Figs. 3, we compare the contributions of the background and the biexciton scattering to the energy shift [insert (a)] and to the absorption [insert (b)] of the upper polariton mode. From this comparison, we infer that the larger effect of the cross Feshbach resonance is arising at a cavity detuning equal to the biexciton binding energy. The weak contribution of the background effect to the absorbance and the position of the resonance centered at a cavity detuning equal to the biexciton binding energy are strong signatures for the cross Feshbach resonance. From the quality of these fits, we conclude that the observed resonance is indeed caused by the scattering of a pair of upper and lower polariton modes into the biexciton state. The fits provide an excellent and quantitative description of the main resonance features across the detuning range, apart from a small deviation on the positive detuning side of the change of absorption. The measured change of absorbance is larger than predicted on the basis of a single scattering channel (the biexciton state) indicating the existence of additional scattering channels above the biexciton energy that correspond to the energy continuum of two-exciton states [39], which sets in at zero detuning.

The dynamics of the cross Feshbach resonance are investigated by measuring the time-integrated transmission spectrum of the delayed probe pulse with respect to the arrival of the pump pulse. In Fig. 4, we display the probe spectrum centered on the upper polariton peak as a function of the pump-probe delay, for a detuning of $\delta = -1.2$ meV, in the vicinity of the cross Feshbach resonance. We observe an energy shift of the upper polariton peak and a reduction of its amplitude around a zero delay, which characterizes the cross Feshbach resonance. The dynamics are clearly revealed by the dependence of the energy shift with delay. The energy shift reaches its maximum value at zero delay: this corresponds to the largest scattering rate of an upper polariton with a lower polariton to the biexciton state, occurring as expected when the optical pulses have the largest temporal overlap. The energy shift varies more rapidly at negative delays than it does at positive delays. The dynamics of the signal at negative delays is governed by the decoherence rate of the upper polariton polarization generated by the probe pulse, while the dynamics at positive delays are governed by the lifetime of the lower polariton population generated by the

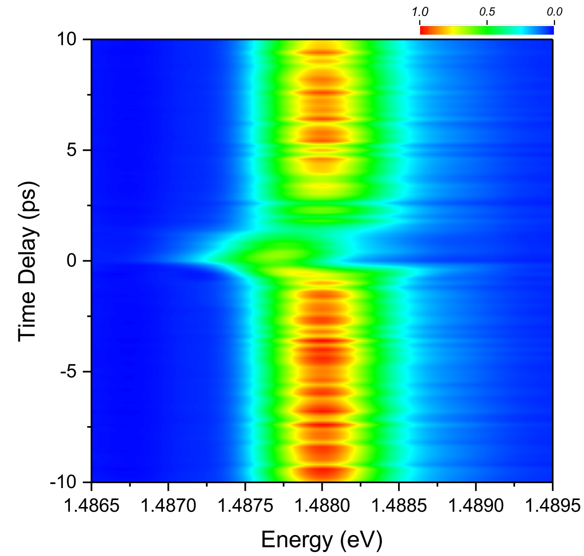

FIG. 4. Dynamics of the cross Feshbach resonance. Transmitted probe spectra around the upper polariton energy (1.488 eV) as a function of pump-probe delay at $\delta = -1.2$ meV. The signal intensity fluctuations along the time delay correspond to shot noise.

pump. This lifetime is determined in part by the scattering rate between a lower and an upper polariton with opposite spins to the biexciton and, for the other part, by the escape rate of the photon from the microcavity.

From the observation of the cross Feshbach resonance, we infer that the generation of entangled photon pairs in semiconductor microcavities could be very effective. Indeed, the efficiency of the generation process of a pair of entangled photons with opposite momenta and spins is determined in part by the rate of the scattering process that generates a biexciton and, in the other part, by the dissociation rate of the biexciton into two outgoing polaritons with opposite spins. We found that about 35% of the transmitted probe photons were converted into biexcitons at the cross Feshbach resonance. Assuming that the dissociation of the biexcitons is about equally distributed between pairs of lower polaritons and pairs of interface polaritons [40], we estimate that one half of the excited biexcitons lead to outgoing pairs of entangled lower polaritons. The flux of outgoing entangled photon pairs is further reduced by the square of the radiative efficiency of the lower polariton mode having a large momentum ($2.6\ \mu\mathrm{m}^{-1}$), which amounts to 7% when accounting for the nonradiative scattering of polaritons by acoustic phonons [41]. At the cross Feshbach resonance, an efficient generation of photon pairs entangled in momentum and polarization is then expected. If we reverse the generation scheme by resonantly exciting the lower polariton modes at large momenta, one would produce pairs of outgoing upper and lower polaritons that are entangled in energy and polarization. The main drawback of this scheme lies in a strongly reduced dissociation rate of the biexciton into pairs of upper and lower polaritons, which is predicted to be smaller by two orders of magnitude [40] in comparison to the rate of dissociation into pairs of lower polariton modes. This reduction is partially compensated by a higher radiative efficiency of a pair of lower and upper polaritons (about 50%). In view of realizing a bright source of entangled photons, a high radiative efficiency will also contribute to enhance the visibility of the photon pairs correlation as it directly depends on the incoherent background emission from the polariton population in the lower branch. Ultimately, this might favor the generation scheme based on a pair of photons entangled in energy and polarization. The polaritonic cross Feshbach resonance is also envisioned to lead to novel studies of many-body correlations using the Bose polaron in an exciton-polariton system [42].

The authors thank B. Deveaud for fruitful discussions.

[*]morteza.navadehtoupchi@epfl.ch

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