Polaritonic Feshbach resonance

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I The effects of decoherence on polariton Feshbach resonance

Feshbach resonance and pump-probe delay dependence

In order to show that the polaritonic Feshbach resonance only persists during the polariton coherence time and disappears with the decoherence of the system, we performed the experiments as a function of the delay between pump and probe pulses. Positive (negative) delay means that the pump (probe) arrives first.

In Figure S1 below we show the energy shift (upper panel) and absorption (lower panel) of the polariton resonance as a function of cavity detuning for different delays. The results obtained at zero, negative and positive delays are shown together to highlight the disappearance of the Feshbach resonance with decoherence at positive delay and its gradual decrease in coherent regime at negative delay.

Let us now explain in detail our experiments and our findings:
In the negative delay configuration, the pump-probe signal only exists for delays within the coherence time of the system. Then the pump-probe signal decays together with coherence and therefore with delay. It is important to point out the significance of the experiments at negative delays, in which we only probe the coherent effects.

We plot in Figure S1a the energy shift of the polariton resonance as a function of cavity detuning for zero, -1.5 ps and -3 ps negative delays. The resonance behaviour clearly appears for zero delay, as the result already presented in Figure 2a in the manuscript. As expected the amplitude of the shifts decreases with the increase of the negative delay. In Figure S1b, we plot the absorption strength of the polariton resonance as function of cavity detuning at zero, -1.5 ps and -3 ps negative delays. Here also we clearly see the decrease of the signal when the delay is increased. However it is important to point out that the resonance features (although decaying) are always present.

With these experiments we demonstrate that the polaritonic Feshbach resonance persists during the coherence time of the polariton system.

At positive delays the pump pulse arrives first and the pump-probe signal exists during the lifetime of the population even if the system loses its coherence. Therefore, in this configuration, the pump-probe signal exhibit features related to coherent and incoherent effects.

We plot in Figure S1c the polariton resonance energy shift and in Figure S1d the absorption as a function of cavity detuning obtained at zero, 1.5 ps and 3 ps positive delays. We show the polaritonic Feshbach resonance features at zero delay in which polaritons are in full coherent regime.
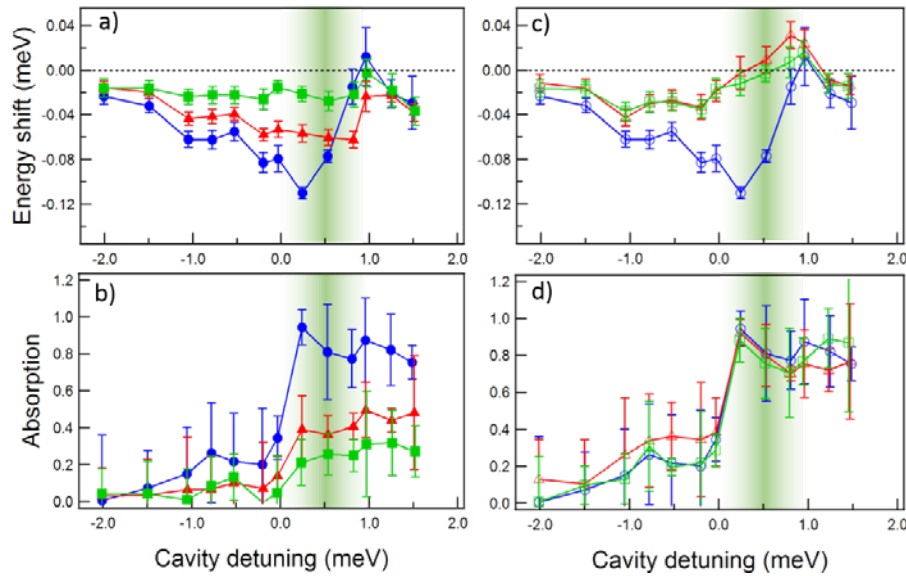
Figure S1 Energy shifts (upper panel) and absorption $ln(P_{ref}/P_{pump-probe})$ (lower panel) of the lower polariton resonance as a function of cavity detuning for different pump-probe delays. Zero, 1.5 ps and 3ps negative (positive) delays correspond respectively to circle, triangle and square full (empty) markers. P_{ref} and $P_{pump-probe}$ respectively represent the amplitude of lower polariton resonance without and with pump excitation. Feshbach resonance in full coherent regime at zero delay. (a and b) Negative pump-probe delays and the persistence of Feshbach resonance in coherent regime: The signal decreases with coherence and therefore with delay. (c and d) Positive pump-probe delays and the disappearance of Feshbach resonance in incoherent regime: The energy shift resonance feature disappears with decoherence therefore with delay. However the absorption persists in incoherent regime due to the presence of polariton population.

Upon increasing the delay, incoherent effects take place bringing a destruction of the resonance feature. Indeed the dispersive shape energy shift, characteristic of the Feshbach resonance is definitely washed out. Moreover, contrary to the results at negative delays the absorption variation of the polariton resonance persists for positive delays due to the presence of the polariton population.

As a conclusion, measurements performed at negative and positive delays give an unambiguous proof of Feshbach resonance as a coherent effect:

(i)   The Feshbach resonance characteristic features both for the energy shift and the amplitude of the absorption evolve with coherence at negative delay.

(ii)   Decoherence kills Feshbach resonance. This is clearly demonstrated with the disappearance of the resonance feature in the energy shift for positive delays. Because a real population is created at positive delays, the absorption persists manifesting the presence of incoherent population effects.

Both results together evidence that incoherence prevents polaritonic Feshbach resonance.

II Background interactions of anti-parallel spin polaritons

There are several possible interaction mechanisms that may contribute to the background interactions. We can mention indirect scattering through dark exciton states [26, 31], and also through excited exciton states [32], exciton-exciton scattering continuum correlations [33], which are attractive. Repulsive interactions such as Van-der-Waals and electrostatic interactions [26] could also contribute. However it is very difficult to determine their relative contributions in the overall energy shift as shown by the different models that can be found in the literature [26, 31, 32, 33, and references therein].

We use a phenomenological model in which the background interaction and the biexciton-exciton interaction are two independent constants. Our experimental observations (energy shift and absorption) can only be reproduced using a background interaction constant with a fixed negative value without changing other parameters except the density of polaritons (Figure 2 and 3 in the paper). In addition, neither null nor positive background constant allow to reproduce our data even by trying to adjust the value of the biexciton-exciton coupling g_{BX} constant.

Here background interaction and biexciton-exciton interaction play a very different role to the Feshbach effect. The biexciton-exciton interaction allows to reproduce the two main effects of the Feshbach resonance namely the strong absorption and the change of the magnitude and of the sign of the energy shift (see Figure S2). The background interaction only acts as an offset of the energy shift – which is negative - as illustrated on Figure S2.
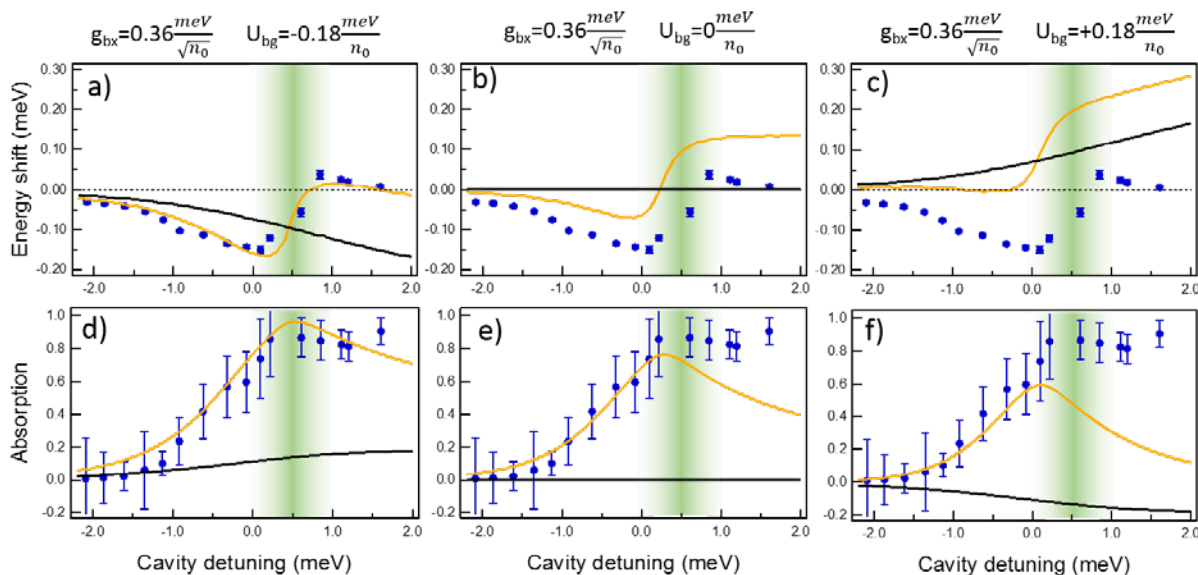

Figure S2 Effect of the background interaction constant as a fitting parameter on data shown in Figure 2 (in the paper). *Upper panel* Energy shifts of the pump-probe spectrum and *lower panel*, $ln(P_{ref}/P_{pump-probe})$ as a function of cavity detuning. Blue circles are experimental results, orange and black lines stand for numerical simulations with and without polariton-biexciton coupling respectively. (a,d), (b,e) and (c,f) show the effect of different values of U_{bg} on the orange fitting curve.

III Origin of the coupling term in the Feshbach resonance

Different transitions between polariton and biexciton could be observed through pump/probe experiments. First, the direct transition from a polariton to the biexciton state as reported by Borri *et al.* [19]. Second, the direct coupling between an exciton and a cavity photon to reach the biexciton state. This process is at the origin of the oscillator strength transfer from excitons to biexcitons reported in [20].

Setting the two anti-parallel polariton spin state in resonance with the biexciton gives rise to the polaritonic Feshbach resonance. Our Feshbach resonance observation is defined in the framework of the "bipolariton model" [23, 24]. Note that in our model, the polariton-biexciton coupling constant g_{BX} describes the anti-parallel spin exciton+exciton coupling to biexciton. This effect is induced by the Coulomb interaction of excitons. The Feshbach resonance could also be described in the framework of another model the "giant oscillator strength model" [24, 18]. In this model the coupling term is Ω_B, which refers to the photon+exciton coupling to biexciton.

To highlight all pathways allowing to couple to biexcitons, we have included in our "bipolariton model", in addition to the g_{BX} coupling, the coupling term Ω_B. This model Hamiltonian is then rewritten as:

$$H = \Omega_X(a_{c\downarrow}\psi_{X\downarrow}^+ + a_{c\downarrow}^+\psi_{X\downarrow}) + \Omega_B(a_{c\downarrow}\psi_{X\uparrow}\psi_{BX}^+ + a_{c\downarrow}^+\psi_{X\uparrow}^+\psi_{BX} + a_{c\uparrow}\psi_{X\downarrow}\psi_{BX}^+ + a_{c\uparrow}^+\psi_{X\downarrow}^+\psi_{BX}) + \frac{U_{bg}}{2}\left(\psi_{X\uparrow}^+\psi_{X\downarrow}^+\psi_{X\downarrow}\psi_{X\uparrow}\right) + g_{BX}(e^{-i\theta}\psi_{BX}^+\psi_{X\uparrow}\psi_{X\downarrow} + e^{i\theta}\psi_{BX}\psi_{X\uparrow}^+\psi_{X\downarrow}^+)$$

In terms of the 3*3 matrix, after mean field approximation the Hamiltonian is the following:

$$H_{eff} = \begin{bmatrix} \psi_{X\downarrow} & a_{c\downarrow} & \psi_B \end{bmatrix}^+ \begin{bmatrix} \varepsilon_X + U_{bg}n_{X\uparrow} - i\frac{\gamma_X}{2} & \Omega_X & g_{BX}\exp(i\theta)\sqrt{n_{X\uparrow}} - \Omega_B\sqrt{n_{c\uparrow}} \\ \Omega_X & \varepsilon_C - i\frac{\gamma_c}{2} & \Omega_B\sqrt{n_{X\uparrow}} \\ g_{BX}\exp(-i\theta)\sqrt{n_{X\uparrow}} - \Omega_B\sqrt{n_{c\uparrow}} & \Omega_B\sqrt{n_{X\uparrow}} & \varepsilon_B \end{bmatrix} \begin{bmatrix} \psi_{X\downarrow} \\ a_{c\downarrow} \\ \psi_B \end{bmatrix}$$

Here $n_{C\uparrow}$ ($n_{x\uparrow}$) corresponds to the photonic (excitonic) density generated by the pump beam. In addition, we have introduced a phase term between the two couplings g_{BX} and Ω_B required to generalize the model.

Let us now detail the respective significance of the couplings g_{BX} and Ω_B in the Feshbach resonance process.

First, in Figure S3a and b (Figure 2a and b in the paper), we show that the experimental results are very well reproduced by our model, when we set $\Omega_B=0$ and consider the g_{BX} coupling as the unique active mechanism in the polaritonic Feshbach resonance. Second, in Figure S3c and d, we now set $g_{BX}=0$ and then we present the results of model when the Ω_B coupling term is the only responsible for the effect. This model also reproduces the experimental results reasonably well at resonance, but the shape of the absorption feature deviates for both very negative and positive detuning. These results enlighten that indeed both terms are giving rise to a Feshbach resonance, however our experimental observation favors the Coulomb term g_{BX} coupling over the giant oscillator coupling.
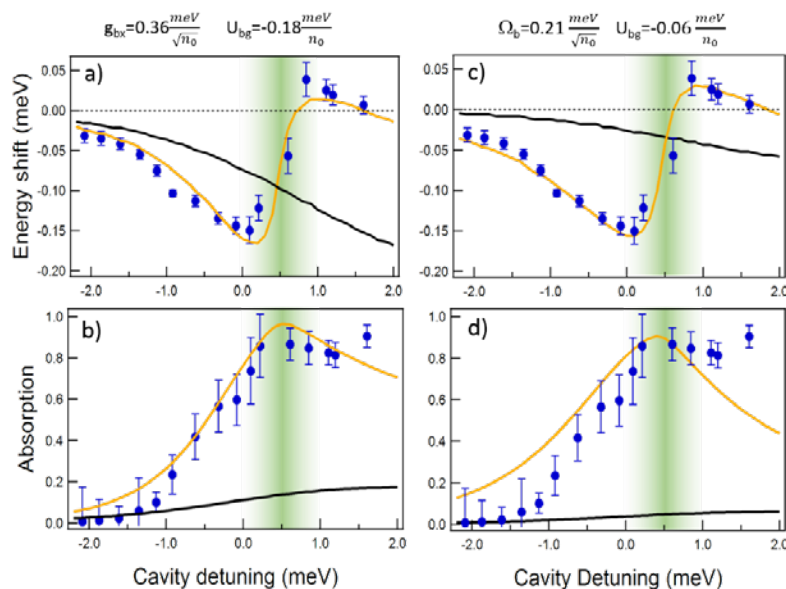
Figure S3 Comparison between the g_{BX} exciton+exciton to biexciton and Ω_B exciton+ photon to biexciton processes. Both coupling terms reproduce the resonance features. All other parameters are same as in the paper.

In order to examine more precisely the model, we now compare our experimental results with the theoretical curves obtained by using the same value for both coupling terms g_{BX} and Ω_B (Figure S4), we only change the phase term θ. In Figure S4a it is clearly shown that, when setting $\theta=0$, both pathways cancel, and no resonance is observed in the theoretical results. Conversely, for phases set either to $\pi/2$ and π the theory plots recover the Feshbach resonance (Figures S4b and c). However, again it deviates for both negative and positive detuning in the absorption feature due to the contribution of the Ω_B coupling.
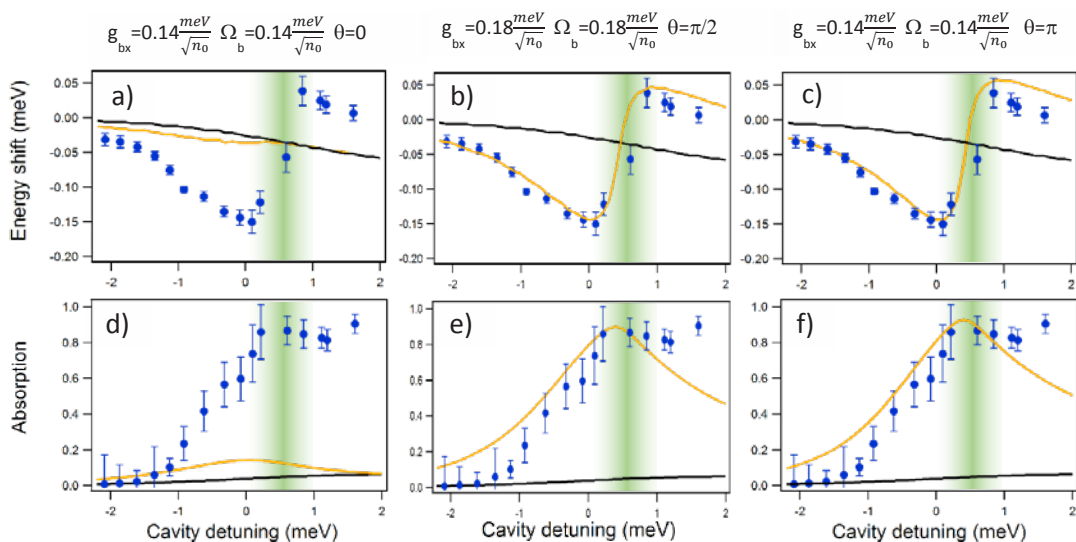

Figure S4 Same values for the g_{BX} exciton+exciton to biexciton and exciton+photon to biexciton (Ω_B) processes and different phases. The resonance feature can be reproduced for phases differing from zero. All other parameters are same as in the paper except $U_{bg}=-0.06\frac{meV}{n_0}$.

This general model enlightens that different combination of parameters and phase relations between the coupling terms are reasonably able to describe the resonance features. However, as the phase value cannot be determined a priori within this generalized model, we cannot obtain, with the limited information available, a precise determination of all parameters. In particular, each term alone gives a reasonable account of the experimental results. The lesser quality of the fit when considering only the polariton-biexciton coupling through the term Ω_B, although it gives rise to a nice Feshbach resonance, brings us to favor the bipolariton model with the Coulomb term g_{BX} as unique polariton-biexciton coupling. This gives us the best fit to our Feshbach resonance results.

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