Exciton-polaritons gas as a nonequilibrium coolant

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Using angle-resolved Raman spectroscopy, we show that a resonantly excited ground-state exciton-polariton fluid behaves like a nonequilibrium coolant for its host solid-state semiconductor microcavity. With this optical technique, we obtain a detailed measurement of the thermal fluxes generated by the pumped polaritons. We thus find a maximum cooling power for a cryostat temperature of 50K and below where optical cooling is usually suppressed, and we identify the participation of an ultrafast cooling mechanism. We also show that the nonequilibrium character of polaritons constitutes an unexpected resource: each scattering event can remove more heat from the solid than would be normally allowed using a thermal fluid with normal internal equilibration.

In this work, instead of bare photons, we consider using resonant optical pumping of ground state exciton-polaritons to excite polariton ASF, and thus generate a cooling power in a solid-state semiconductor microcavity (MC) in the strong coupling regime (cf. Fig. 1a). Polaritons are bi-dimensional quasi-particles that benefit from a half-photonic, half-excitonic nature, to achieve much stronger interaction with phonons than photons [16, 17]. Fig. 1c shows the principle of the most interesting cooling mechanism achieved by this excitation scheme, and that we report in this letter: within a short lifetime of 1 ps, a polariton can be excited into a higher energy state by absorbing a thermal acoustic phonon of average energy \( \hbar \Omega_f \). Its subsequent radiative recombination results in the net energy transfer of \( \hbar \Omega_f \) from the thermal phonon bath to the outside electromagnetic vacuum. Two additional thermal exchange mechanisms also take place, a cooling one shown in Fig. 1d, and a heating one shown in Fig. 1e, as will be discussed later.

Polaritons have many advantages over other optical methods to cool down semiconductor materials: their cavity-like dispersion (cf. \( \omega(k) \)) in Figs. 1c-e fully inhibits the Stokes emission which is a source of phonon emission. Moreover, owing to their bi-dimensional degree of freedom, a 2D continuum of states is available for anti-Stokes scattering, so that thermal phonons of ar-
The polariton field absorbs thermal phonons resulting in polariton ASF emission. The wavy arrows depict photons, while the spring-like arrows represent phonons.

Phonon energy is counted positive (negative) for emission (absorption). Fast cooling mechanism b) removes phonons of average energy $\hbar\Omega_1$, Slow cooling mechanism c) removes phonons of average energy $\hbar\Omega_2$, while two-photon absorption d) generates a phonon cascade (heat) of average energy $\hbar\omega_0 - \hbar\Omega_2$. The gray rectangle is the area outside the polariton light cone.

Another useful property of polaritons in this context is their ultra-light mass of $\hbar$ at focused in a spatially Fourier-transform spot of a phonon cascade (heat) of average energy $\hbar\omega_0$.

Finally, since ASF emission is isotropic, the total ASF anti-Stokes photon emission rate in counts per seconds.

Details on the experimental setup and data treatment are given in the supplemental materials [18].
A state (Fig. 1d). Within a spectral analysis of timescale of \( \sim \) we call \( \hbar \) cooling power, and involves two distinct mechanisms. In fluorescence at higher energy. It thus provides the wanted ground state polaritons of energy \( P \).

Experimentally measured non-resonant spectrum its contribution is maximum at low energy (only the high laser power \( P_{\text{las}} \) ). (i) the fact that for such large \( \Delta \) the phonons involved are of the optical type, and thus exhibit a coupling strength with polaritons fifty times larger than the acoustic ones involved in the fast cooling [6].

We have carried out this analysis at different cryostat temperatures. We found that the fast cooling mechanism has a peak contribution of \( P = 61\% \) at \( T = 20K \), and remains significant up to \( T \sim 100K \) (cf. Fig 3b). This fast cooling mechanism is unique to polaritons: its timescale is fixed by the polariton lifetime which is two orders of magnitude shorter than the slow cooling one. This dynamics is even faster than the phonons typical thermalization time [22]. The observation of this fast cooling mechanism in a MC in the strong coupling regime is the key result of this letter. The slow cooling mechanism is reminiscent from that involved in exciton-enhanced optical cooling [9]. At such low temperatures, its contribution can seem surprising as it involves thermal phonons of energy comparable with \( \Delta \gg k_B T \). However, the weak phonon population at this energy is compensated by two aspects: (i) the excitonic density of states, which is four orders of magnitude larger than that of polaritons, and (ii) the fact that for such large \( \Delta \) the phonons involved are of the optical type, and thus exhibit a coupling strength with polaritons fifty times larger than the acoustic ones involved in the fast cooling [6].

Now that we have a measurement of the three main mechanisms involved in the heat exchange between polariton and thermal phonons, we can derive an experimental value of the thermal energy removed from the MC per units of time (i.e. the cooling power) as

\[
P_{\text{fr}}(P_{\text{las}}, T) = \int d\omega \left\{ P_{\text{las}} h(\omega - \omega_0) A^{(1)}(\omega, T) \\
- P_{\text{las}}^2 h(2\omega_0 - \omega) A^{(2)}(\omega, T) \right\},
\]

where \( h(\omega - \omega_0) \) is the energy of a phonon which has been removed when a photon is detected at the energy \( h\omega \). Positive \( P_{\text{fr}} \) means cooling while negative means heating. Note that in this approach, \( P_{\text{fr}} \) is a lower bound of the true cooling power since due to the objective finite numerical aperture, only 46% of the whole momentum space is accessible. Indeed, the inaccessible large \( k_{||} \) ring involves the absorption of high energy thermal phonons, that have a large contribution to cooling via the
The maximum cooling power $p_f$ and the cooling power $P_{\text{tot}}$ could be largely increased by making two fairly simple changes in the MC design. Firstly, by lowering the quality factor $Q = \omega_0/(2\pi\gamma)$ (presently, $Q = 5600$), the ratio of two-photon absorption rate over cooling rate would decrease since the earlier scales like $1/\gamma^2$ and the latter scales like $1/\gamma$. Obviously, $Q$ cannot be decreased to arbitrarily low values as it must remain large enough for the strong coupling regime to be preserved. Secondly, when $\Delta$ exceeds the energy $\hbar\Omega_{\text{max}}$ of the highest frequency optical phonon mode, the scattering of a polariton into the excitonic reservoir by a single phonon becomes forbidden by energy conservation.

A strong suppression of the slow cooling mechanism in favor of the fast one is thus achieved. Such a suppression has been demonstrated already in a different context in a ZnO MC [24]. An additional condition required to preserve the fast cooling mechanism is that the excitonic fraction should remain significant. Both conditions are easily met in state-of-the-art Selenide and Telluride microcavities that combine large Rabi splitting and low LO phonon energy.

This work also gives us a striking insight on the thermal properties of a nonequilibrium cryogenic fluid (polaritons) interacting with a thermal bath (phonons) over a timescale too short for it to thermalize. The thermodynamical point-of-view on this experiment is summarized in Fig. 3. Polaritons are injected with an effective temperature much colder than that of thermal phonons (the pump laser injects polaritons in their ground state). During the cooling mechanisms, polaritons pick up heat from the thermal phonon bath, and then recombine radiatively. The steady-state polariton gas resulting from this interaction is highly non-thermal: it consists of two independent components: a "cold" gas that did not interact with phonons and recombines at the same energy at which it entered the MC, and a smaller "hot" one, in which the captured heat is distributed according to a non-thermal distribution function, a measurement of which is given by $A^{(1)}(\omega)$. This "hot" and "cold" components do not mix up like in a normal fluid, because at the low densities involved in this cooling scheme, polaritons do not interact with each others. The properties of this "hot" component are rather unusual: in Fig. 3c we plotted $\hbar\Omega_c(T)$ the average energy of $A^{(1)}(\omega)$, that represents the average energy removed from the solid per scattering event. $\hbar\Omega_{\text{eq}}(T)$, the average energy of an hypothetical polariton gas at thermal equilibrium is shown on the same plot. We find that between $T = 4.2$K and $T = 100$K $\hbar\Omega_c > \hbar\Omega_{\text{eq}}$, meaning that the thermal energy removed from the solid by each polariton of the "hot" component is larger than if they were at thermal equilibrium with the lattice. We can also compare $\hbar\Omega_c(T)$ with $\hbar\Omega_{\text{ph}}(T) \approx 2.701k_bT$, the average energy of the thermal phonon bath. It appears that between $T = 4.2$K and $T = 50$K, $\hbar\Omega_c > \hbar\Omega_{\text{ph}}$, meaning this time that the "hot" component of the polariton gas is even "hotter" than the slow channel. We extracted $P_{f}$ for temperatures ranging from $T = 4.2$K to $T = 150$K. The result is summarized in Fig. 3a versus temperature and laser power. We find that the maximum cooling power $P_{f}^{\text{max}} = (0.10 \pm 0.02)\mu$W, with $\rho = 40\%$ is achieved at $T = 50$K, and remains positive below. As explained above, this is only possible because polariton ASF does not involve a discontinuous electronic density of states but rather a continuous one with no energy gap between the pump polaritons and the available anti-Stoke states. Therefore, phonons of very low energy can be pumped out from the thermal bath by this method. Such a behavior is another unique feature of polariton cooling and constitutes the second key result of this letter. Note that this cooling power is generated within a micron-scale volume resulting in a large cooling power density of $p_f = (80 \pm 16)\mu$W.cm$^{-3}$. Finally, we see in Fig. 3c (red diamond) why the main limitation to polariton cooling is two-photon absorption, indeed although this absorption rate is low as compared to the cooling rate (at least at power $P_{\text{las}}$ low enough), each such event releases a large amount of heat $h(\omega_0 - \Omega_x)$ in average, ranging from 2760 meV to 2780 meV.
phonon bath itself. This surprising result suggests that introducing a nonequilibrium character to a coolant fluid might constitute a useful resource to enhance its performance.

In this work we have shown, as an experimental proof-of-principle, that polaritonic cooling of a semiconductor MC works and presents unusual properties with respect to state-of-the-art methods of optical cooling in solids. Firstly, owing to the specific polaritonic density of states, that allows removing low energy thermal acoustic phonons, cooling can be achieved at arbitrarily low cryostat temperature. Secondly, polaritonic cooling involves a new mechanism referred to as fast cooling that is unique to polaritons, and opens up an experimental window on nonequilibrium thermodynamics of phonons. Finally, we have shown that polariton fluids constitute an experimental model system to study the heat transport properties between a thermal bath and a nonequilibrium fluid.

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[1] Special issue on "Laser Cooling and Trapping of Atoms", edited by S. Chu and C. Wieman, J. Opt. Soc. Am. B 6, 2020-2270 (1989).
[2] K. C. Neuman and S. M. Block, Review Of Scientific Instruments, 75, 2787 (2004).
[3] S. Gigan, H. R. Böhm, M. Paternostro, F. Blaser, G. Langer, J. B. Hertzberg, K. C. Schwab, D. Bäuerle, M. Aspelmeyer and A. Zeilinger, Nature 444, 67 (2006).
[4] O. Arcizet, P. F. Cohadon, T. Briant, M. Pinard and A. Heidmann, Nature 444, 71 (2006).
[5] J. Chan, T. Alegre, A. Safavi-Naeini, J. Hill, A. Krause, S. Groblacher, M. Aspelmeyer and O. Painter, Nature 478, 89 (2011).
[6] P. Pringsheim, Z. Phys. A 57, 739-746 (1929).
[7] M. Sheik-Bahae and R. I. Epstein, Nature Photon. 1, 693-699 (2007).
[8] D. V. Seletskiy, S. D. Melgaard, R. I. Epstein, A. Di Lieto, M. Tonelli and M. Sheik-Bahae, Optics Express, 19, 18229 (2011)
[9] G. Rupper, N. H. Kwong and R. Binder, Phys. Rev. Lett. 97, 117401 (2006).
[10] M. Sheik-Bahae and R. I. Epstein, Phys. Rev. Lett. 92, 247403 (2004).
[11] H. Gauck, T. H. Groerer, M. J. Renn, E. A. Cornell and K. A. Bertness, Appl. Phys. A 64, 143-147 (1997).
[12] E. Finkeissm, M. Potemski, P. Wyder, L. Vina and G. Weimann, Appl. Phys. Lett. 75, 1258-1260 (1999).
[13] J. Zhang, D. Li, R. Chen and Q. Xiong, Nature 493, 504-508 (2013).
[14] D. V. Lang and C. H. Henry, Phys. Rev. Lett. 35 1525 (1975).
[15] C. Weisbuch, M. Nishiokas, A. Ishikawa, and Y. Arakawa Phys. Rev. Lett. 69, 3314 (1992).
[16] A. Fainstein, B. Jusserand and V. Thierry-Mieg, Phys. Rev. Lett. 78, 1576 (1997).
[17] G. Rozas, A. E. Bruchhausen, A. Fainstein, B. Jusserand, and A. Lemaître, arXiv:1405.0880 (2014).
[18] See supplemental material for details.
[19] T. Amand and X. Marie, Exciton Spin Dynamics in Semiconductor Quantum Wells in Spin Physics in Semiconductors edited by M. I. Dyakonov, Springer Berlin Heidelberg (2008), ISBN 978-3-540-78820-1.
[20] K. Sebald, M. Seyfried, S. Klembt, S. Bley, A. Rosnauer, D. Hommel and C. Kruse, Appl. Phys. Lett. 100, 161104 (2012).
[21] M. Umlauff, J. Hoffmann, H. Kalt, W. Langbein, J. M. Hvam, M. Scholl, J. Sollner, M. Henken, B. Jobst and D. Hommel, Physical Review B, 57 1390 (1998).
[22] G. Rozas, M. F. P. Winter, B. Jusserand, A. Fainstein, B. Perrin, E. Senenova and A. Lemaître, Phys. Rev. Lett. 102, 015502 (2009).
[23] S. Rudin, T. L. Reinecke and B. Segall, Phys. Rev. B, 42 11218 (1990).
[24] A. Trichet, L. Sun, G. Pavlovic, N.A. Gippius, G. Malpuech, W. Xie, Z. Chen, M. Richard and Le Si Dang, Phys. Rev. B 83, 041302(R) (2011).

SUPPLEMENTAL MATERIAL FOR: Exciton-polaritons gas as a nonequilibrium coolant

Experimental setup and Numerical data treatment

The raw polariton ASF data are acquired and treated in the following way.

• The sample is placed in vacuum inside a variable temperature cryostat used to set the MC base temperature. it is mounted on its holder with an upward angle of 13° with respect to the excitation/detection objective optical axis. In this way, spurious reflection of the laser on the objective lenses are suppressed upon exciting at $k_{\|}=0$, and more importantly, ASF detection is possible up to an emission angle of 43° (the microscope objective has a numerical aperture of 0.5). The CW laser beam is injected at a well defined incidence angle on the MC by focusing it with a $f = 300$mm lens onto the input Fourier plane of the objective, so that a spatially Fourier transform excitation spot is achieved, with a diameter of 20µm on the surface of the sample and an angular
spread of $3^\circ$. The laser beam is tuned, angle-wise and in wavelength at resonance with the polariton ground state ($k_{\parallel} = 0, \omega_0$), where $\hbar \omega_0 = 2790$ meV.

- The excitation laser and the detection are cross-polarized by Glan-Thompson polarizers, thus achieving a rejection efficiency of $10^{-7}$. However, while the laser is fully suppressed with respect to the ASF intensity, a bright cross-polarized polariton emission occurs from the ground state, likely due to a weak local spin-anisotropic disorder. This resonant photoluminescence is three orders of magnitude weaker than the incoming laser but still three to four orders of magnitude brighter than the ASF. To further reject this signal, we use two consecutive filtering steps: the emission Fourier plane is imaged on the entrance slit of the monochromator. It is shifted by $80 \mu m$ with respect to $k_{\parallel} = 0$ such that the emission at $k_{\parallel} \leq 0.58 \mu m^{-1}$ is blocked. The remaining spurious resonant signal entering the monochromator is then shifted away spectrally by $1.65$ meV with respect to the bottom of the polariton dispersion, from the edge of the charged coupled device (CCD) sensitive area. Overall, these filtering steps reject emission angles between $0^\circ$ and $8.4^\circ$, while ASF is measured from $8^\circ$ to $43^\circ$.

- Absolute calibration of the setup transmission is carried out in the following way: a calibrated silicon photodetector is placed on the transmitted port of the last beamsplitter, while the reflected port leads to the microscope objective. We thus obtain an accurate reading of the laser power $P_1$ at this point of the setup. The power actually entering the MC is $P_{\text{las}} = P_1 T_{\text{obj}} T_{\text{pol}}$, where $T_{\text{obj}} = 0.75$ is the objective transmission at $\hbar \omega_0$ and $T_{\text{pol}} \simeq 1 - R_{\text{pol}}$ is the measured MC transmission at resonance. Then the optical efficiency of the setup is calibrated using a reflection of the laser on the MC at a wavelength redshifted from the polariton resonance where it behaves as a $\sim 100\%$ mirror. Using our calibrated photodetector, when a photon is emitted by the MC within the detected emission cone, it has a probability of $1.11\%$ to be detected on the CCD. In other terms, at this wavelength, a conversion efficiency of $2.48 \times 10^{10}$ counts/s is found per $\mu W$ of fluorescence.

- The image thus obtained on the CCD is a measurement of polariton ASF with spectral (Horizontal axis of the CCD) and angular resolution (vertical axis). The function converting the vertical pixels into true wavevector $(k_x, k_y)$ is known by a previous CCD-pixel-to-angle calibration of the microscope objective. The CCD image is thus reshaped according to this function and results in image like that of Fig.2.a (main text). In order to get the polariton ASF count rate $I_{\text{ASF}}$, the lower polariton branch is fitted with the theoretical one (obtained from the usual two coupled harmonic oscillators model). From that fit, a mask is created that borders the raw data points $I_{\text{ASF}}(\omega_n, P_{\text{las}})$ are fitted with the function $A^{(1)}(\omega_n) P_{\text{las}} + A^{(2)}(\omega_n) P_{\text{las}}^2$, where $A^{(2)}(\omega_n)$ and $A^{(2)}(\omega_n)$ are the fitting parameters. The thus obtained spectral densities $A^{(1)}(\omega)$ and $A^{(2)}(\omega)$ characterizes the ASF response to optical excitation in counts/s/meV$\mu W$ and counts/s/meV$\mu W^2$ respectively (cf. Fig.2.c of the main text). The error bars are obtained from the $95\%$ confidence bound of this fitting procedure. This method allows an accurate separation of the two-photon absorption related source of heat, from the source of cooling, namely the fast and slow cooling mechanisms depicted in Fig.1.a and Fig.1.b of the main text.

- For a given temperature, the fractions of the ASF spectrum that behave linearly $A^{(1)}(\omega)$ and quadratically $A^{(2)}(\omega)$ with respect to $P_{\text{las}}$ are obtained from the dataset $I_{\text{ASF}}(\omega, P_{\text{las}})$: for each energy pixel $\omega_n$ the data points $I_{\text{ASF}}(\omega_n, P_{\text{las}})$ are fitted with the function $A^{(1)}(\omega_n) P_{\text{las}} + A^{(2)}(\omega_n) P_{\text{las}}^2$, where $A^{(2)}(\omega_n)$ and $A^{(2)}(\omega_n)$ are the fitting parameters. The thus obtained spectral densities $A^{(1)}(\omega)$ and $A^{(2)}(\omega)$ characterizes the ASF response to optical excitation in counts/s/meV$\mu W$ and counts/s/meV$\mu W^2$ respectively (cf. Fig.2.c of the main text). The error bars are obtained from the $95\%$ confidence bound of this fitting procedure. This method allows an accurate separation of the two-photon absorption related source of heat, from the source of cooling, namely the fast and slow cooling mechanisms depicted in Fig.1.a and Fig.1.b of the main text.

- Finally to separate the fast from the slow cooling contribution in the linear part of the ASF spectrum $A^{(1)}(\omega)$, we take advantage of the fact that both mechanisms lead to very different spectra, in particular at this detuning where the non-resonant polariton emission (as expected for the slow cooling fraction of the ASF) presents a strong bottleneck, i.e. an intensity maximum high above the ground state ($> 15 meV$ in our case). Indeed the fast cooling fraction of the ASF is on the contrary peaked very close to the ground state (at about $0.8 meV$) and then decays quasi-exponentially with energy. Thus in order to extract the fraction $\rho$ of fast cooling in $A^{(1)}(\omega)$, the latter is fitted with the weighted sum of the non-resonant photoluminescence spectrum $I_{\text{PL}}(\omega)$ obtained by exciting the MC with a CW non-resonant laser at $2990 meV$, and the ASF theoretical spectrum due to the fast cooling mechanism $I_{\text{th}}(\omega)$. In other words, $A^{(1)}(\omega) = C \rho I_{\text{th}}(\omega) + C(1-\rho)I_{\text{PL}}(\omega)$, where $C$ is a constant.

- The average phonon energy absorbed by the fast cooling process reads

$$\hbar \Omega_f = \frac{\sum_n \hbar(\omega_n - \omega_0) I_{\text{th}}(\omega_n)}{\sum_n I_{\text{th}}(\omega_n)}$$

(A.2)
The average phonon energy absorbed by the slow cooling process reads
\[
\hbar \Omega_s = \frac{\sum_n \hbar(\omega_n - \omega_0) I_{PL}(\omega_n)}{\sum_n I_{PL}(\omega_n)}
\]  \hspace{1cm} (A.3)

The average phonon energy absorbed by both cooling processes reads
\[
\hbar \Omega_c = \frac{\sum_n \hbar(\omega_n - \omega_0) A^{(1)}(\omega_n)}{\sum_n A^{(1)}(\omega_n)}
\]  \hspace{1cm} (A.4)

The average phonon cascade energy emitted after a two-photon absorption process reads
\[
\hbar \Omega_2 = \frac{\sum_n \hbar(2\omega_0 - \omega_n) A^{(2)}(\omega_n)}{\sum_n A^{(2)}(\omega_n)}
\]  \hspace{1cm} (A.5)

and since \( A^{(2)}(\omega) \) has the same shape as \( I_{PL}(\omega) \), \( \hbar \Omega_2 = \hbar(\omega_0 - \Omega_s) \).

**Polarization properties of the polariton anti-Stokes fluorescence**

Our method of rejection of the laser to detect the weak ASF signal relies on a cross-polarized detection scheme. For the slow cooling mechanism and the two-photon absorption related fluorescence, a depolarized emission is expected since reservoir excitons and a fortiori hot free carriers undergo a fast spin scrambling. Therefore a cross-polarized detection scheme allows measuring one half of the counts of such origins. In the fast cooling mechanism, a pump polariton is scattered into an excited polariton by absorption of a single thermal phonon. In principle, in this inelastic scattering process, polariton spin flip is strongly suppressed as compared to bare excitons. The reason is the following: acoustic phonons are lattice deformations that do not interact with the carriers spin. They do not interact either with the electron orbital momentum since the latter has a s-like symmetry. The hole orbital however, is p-like and lattice acoustic phonons are lattice deformations that do not interact with the carriers spin. They do not interact either with the electron orbital momentum since the latter has a s-like symmetry. The hole orbital however, is p-like and lattice deformations can thus couple different hole orbital states with each other (cf. ref [19] of the main text). As a result, deformations can thus couple different hole orbital states with each other (cf. ref [19] of the main text). As a result,

During their lifetime, anti-Stokes polaritons interact strongly with the MC in-plane disorder. In this case, the depolarized polaritons gets scattered over every directions isotropically (Rayleigh scattering), while linearly polarized polaritons gets depolarized in the process, since this scattering mechanism is also affected by the TE/TM splitting. This additional step thus results in a fully depolarized fast polariton ASF.
This depolarization process of fast ASF is a useful advantage for our experiment since, although we detect cross-polarized with respect to the laser, we miss only about one-half of the emitted photons for any of the cooling or heating mechanisms. Moreover, the isotropic character of the emission allows us to extrapolate the total ASF from an incomplete measurement (a measurement along a single slice in momentum space, defined by the monochromator slit).

**Theoretical model for the fast cooling mechanism**

In order to describe the fast cooling mechanism theoretically we consider a 2D polariton condensed gas immersed in a 3D phonon bath. To determine the polariton transition rate $W_{k-k'}$ from an initial momentum $k$ to a final momentum $k'$ mediated by phonon absorption we use the Fermi golden rule:

$$W_{k-k'} = \frac{2\pi}{\hbar} \sum_{q, q_{\parallel}} |\langle k' | \langle n_{q_{\parallel}, q_z} - 1 | H_{pol-ph} | n_{q_{\parallel}, q_z} \rangle | k \rangle|^2 \delta(E_{pol}(k') - E_{pol}(k) - E_{ph}(q_{\parallel}, q_z))\delta_{k', k + q_{\parallel}}$$  \hspace{1cm} (A.6)

where

$$H_{pol-ph} = \sum_{q_{\parallel}'} \sum_{q_{z}', k', k} X_{q_{\parallel}'} X_{k'} G(q_{\parallel}, q_{\parallel}') \delta_{k', k + q_{\parallel}} (c_{q_{\parallel}', q_{z}'}^\dagger c_{-q_{\parallel}', q_{z}'} - c_{-q_{\parallel}', q_{z}'}^\dagger c_{q_{\parallel}', q_{z}'}^\dagger) b_{k'}^\dagger b_k,$$  \hspace{1cm} (A.7)
with $\lambda_k$'s being the Hopfield coefficients, $b_k^\dagger (b_k)$ is the creation (destruction) operator of 2D exciton with in-plane wave vector $k$. $c_{q^1, q_z}^\dagger (c_{q^1, q_z})$ is the creation (destruction) operator of 3D phonons with wave vector $(q^1, q_z)$, and

$$G(q^1, q_z) = \frac{4}{\sqrt{2\rho \nu}} \left[ |D_e I_{e}^0(|q^1|)I_{e}^z (q_z)| - D_h I_{h}^0(|q^1|)I_{h}^z (q_z)\right]$$

(A.8)

where $u = 3.5 \times 10^3$ m.s$^{-1}$, $\rho = 5.65 \times 10^3$ kg.m$^{-3}$ are the longitudinal sound velocity and the density in ZnSe, and the quantization volume is given by $V$. $D_e = -13.26$ eV and $D_h = -6.56$ eV are the deformation potential for the electron and hole. $I_{e}^0(|q^1|)$ and $I_{h}^0(|q^1|)$ are the overlap integrals between the electron (hole) bound in a 1s quantum well exciton state and the phonon-modes. As derived in [45],

$$I_{e}^0(|q^1|) = \left[ 1 + \left( \frac{m_e(c)}{2M} |q^1| \alpha_B \right) \right]^{-3/2}$$

(A.9)

$$I_{h}^0(|q^1|) = \text{FT}\{f_{e}(h)(z)\}$$

(A.10)

where $m_e = 0.18$ and $m_h = 1.26$ are the electron/hole mass in electron mass units, $M = m_e + m_h$ is the exciton mass and $\alpha_B = 4.1$ nm is the bulk ZnSe exciton Bohr radius. FT{} is the one-dimensional spatial Fourier transform, and $f_{e}(h)$ is the electron (hole) quantum well wavefunction along the confinement axis $z$. The latter is calculated according to our actual quantum well parameters: $L_z = 8$ nm thickness and 310 meV confinement energy. Finally,

$$W_{k \rightarrow k'}^f = \frac{2\pi}{\hbar} \sum_{q^1, q_z} n(q^1, q_z)|\lambda_k^2| |\lambda_{k'}^2| G(q^1, q_z)^2 \delta_{k', k} \delta_{q^1} \delta_{q_z} [E_{pol}^f(k') - E_{pol}^f(k) - \epsilon_{ph}(q^1, q_z)]$$

(A.11)

where $E_{pol}^f(k)$ is the dispersion of noninteracting polaritons, the phonon occupation is given by the Bose-Einstein distribution $n(q^1, q_z) = (\exp[E_{ph}(q^1, q_z)/(k_B T)] - 1)^{-1}$, with $E_{ph} = \hbar \nu \sqrt{q^2_1 + q^2_2}$ the dispersion of acoustic phonons. In order to calculate the ASF spectrum, $k = 0$ is taken as the initial state. We include a finite linewidth for the polariton dispersion so that Eq. (A.11) becomes

$$W_{k \rightarrow k'}^f = \frac{2\pi}{\hbar\pi} \sum_{q^1, q_z} n(q^1, q_z)|\lambda_0^2| |\lambda_{k'}^2| G(q^1, q_z)^2 \delta_{k', k} \delta_{q^1} \frac{\gamma}{[E_{pol}^f(k') - \epsilon_{ph}(q^1, q_z)]^2 + \gamma^2}.$$  

(A.12)

The ZnSe material parameters have been taken from refs. [6, 7].

Orders of magnitude of the pump polariton non-radiative recombination rate

Non-radiative recombination (NRR) is a process where an excitation (a pump polariton in our case) relaxes its total energy in the form of a cascade of phonons, involving localized defect states lying in the material bandgap (cf. Fig[5]). It is thus a source of heat. Owing to their largely dominant population, NRR of pump polaritons is the largest potential source of NRR in our microcavity. To our knowledge, NRR of exciton-polaritons has never been measured so far. Indeed, such a measurement is quite challenging as the NRR lifetime is around 6 orders of magnitude longer than the polariton lifetime in the MC.

In order to estimate $P_{NRR}$, the contribution of pump polariton NRR to the heating power, we make the rough approximation the polariton NRR rate $\gamma_{nr}$ is that of exciton multiplied by the excitonic fraction $X^2$. Then, Within a simple rate equation model, $P_{NRR} = P_{las}X^2 \hbar \omega_0 \gamma_{nr}/\gamma$, where $\gamma$ is the polariton radiative rate. In this approximation, we find that $P_{NRR}$ is equal to $P_{max}$ for $\gamma_{nr}^{-1} \approx 1500$ ns.

To get an order of magnitude to compare with, the above estimation can be compared with the excitonic NRR rate $\gamma_{nr,X}^{-1} = 350$ ns found for excitons in ref. [3], in ZnSe, CdMnSe/CdSe, CdMnSe/MoSe, n-doped quantum wells (QWs) with a thickness of $d = 4.5$ nm, measured at $T=500K$. This lower than $\gamma_{nr}^{-1}$, however, in our microcavity, we are dealing with a different material and structure, and with polaritons instead and excitons. Thus, several qualitative arguments advocate for a smaller $\gamma_{nr}$ by at least a few orders of magnitude:

• The first set of arguments concerns the structural properties: (i) the material of our QWs is binary ZnSe, so that the defect density within the QW is thus much lower than a for a ternary material. (ii) Our QWs have a thickness of $d = 8$ nm and are thus twice thicker. Since most defect leading to NRR are formed at the interface
between the QW and the barrier, an approximately twice lower defect density is expected. (iii) Some of us have carried out a TEM investigations of our MC, including the QWs. The defect density which is found is indeed very low as and sets the state of the art for epitaxial II-VI materials [9]. (iv) The electron donor responsible for the n-doping provides non-negligible density of defect state, possibly contributing to NRR.

- The main argument relies on the polariton mass: in the comparison above, it is assumed that the coupling between a polariton and a defect state is only reduced by a factor $X^2$ as compared to an exciton. However, considering the polariton mass typically four orders of magnitude lighter than the exciton one, and according to the Fermi golden rule, its capture by a point-like defect of a size comparable with the lattice parameters must be reduced by the same order of magnitude.

With this set of arguments in mind, we are quite confident that the actual NRR lifetime $\gamma^{-1}_{nr}$ in our MC exceeds significantly 1500 ns and can thus be reasonably neglected.

[1] W. Langbein, I. Shelykh, D. Solnyshkov, G. Malpuech, Yu. Rubo and A. Kavokin, Phys. Rev. B 75, 075323 (2007).
[2] A. Kavokin, G. Malpuech and M. Glazov, Phys. Rev. Lett. 95, 136601 (2005).
[3] C. Leyder, M. Romanelli, J. P. Karr, E. Giacobino, T. C. H. Liew, M. M. Glazov, A. V. Kavokin, G. Malpuech and A. Bramati, Nat. Phys. 3, 628-631 (2007).
[4] F. Tassone, C. Piermarocchi, V. Savona, A. Quattropani and P. Schwendimann, Phys. Rev. B 53, R7642 (1996).
[5] C. Piermarocchi, F. Tassone, V. Savona, A. Quattropani and P. Schwendimann, Phys. Rev. B 53 15834 (1996).
[6] S. Rudin, T. L. Reinecke and B. Segall, Phys. Rev. B, 42 11218 (1990).
[7] M. Cardona and N. E. Christensen, Phys. Rev. B 35, 6182 (1987).
[8] S. K. Zhang, H. Lu, W. B. Wang, B. B. Das, N. Okoye, M. Tamargo, and R. R. Alfano, Journal of Applied Physics, 101, 023111 (2007).
[9] S. Klembt, K. Frank, G. Qian, T. Klein, A. Rosenauer, D. Hommel and C. Kruse, Journal of Crystal Growth 378 270 (2013).