Supplementary Information



Supplementary Figure S1: Calculated intersubband absorption of sample B. We include the effect of carrier-phonon scattering (cp), carrier-carrier scattering (cc), the diagonal scattering contributions (cp+cc, diag) and all terms combined including the non-diagonal scattering (full). The carrier-carrier and carrier-phonon interactions are non-additive de-phasing mechanisms.

Supplementary Note 1: Theoretical framework

In our model the total Hamiltonian in Coulomb gauge and second quantization is given by

$$H = H_0 + H_{cl} + H_{cc} + H_{cp} , (S1)$$

where the Hamiltonian of the non-interacting Bloch electrons, phonons, and photons is represented by H_0 , carrier-light interaction by H_{cl} , carrier-carrier interaction by H_{cc} , and carrier-phonon interaction by H_{cp} :

$$H_{0} = \sum_{\alpha=1,2} \sum_{k} E_{\alpha,k} c_{\alpha,k}^{\dagger} c_{\alpha,k} + \sum_{Q} \hbar \omega_{LO} d_{Q}^{\dagger} d_{Q} + \sum_{k} \hbar \omega_{cav,k} \left(a_{k}^{\dagger} a_{k} + \frac{1}{2} \right)$$
(S2)
$$H_{cl} = i \hbar \Omega_{R} \sum_{k,q} \left[\left(a_{-k} + a_{k}^{\dagger} \right) c_{1,q}^{\dagger} c_{2,k+q} - \left(a_{-k}^{\dagger} + a_{k} \right) c_{2,k+q}^{\dagger} c_{1,q} \right]$$
(S3)
$$+ \frac{\hbar \Omega_{R}^{2}}{\omega_{21}} \sum_{k} \left(a_{-k} + a_{k}^{\dagger} \right) (a_{k} + a_{-k}^{\dagger})$$

$$H_{cc} = \frac{1}{2} \sum_{\alpha,\beta,\gamma,\delta=1,2} \sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} V_{\boldsymbol{q}}^{\alpha\beta\gamma\delta} c_{\alpha,\boldsymbol{k}+\boldsymbol{q}}^{\dagger} c_{\beta,\boldsymbol{k}'-\boldsymbol{q}}^{\dagger} c_{\delta,\boldsymbol{k}'} c_{\gamma,\boldsymbol{k}}$$
(S4)

$$H_{cp} = \sum_{\alpha,\beta} \sum_{\boldsymbol{k},\boldsymbol{Q}} g_{\boldsymbol{Q}}^{\alpha\beta} c_{\alpha,\boldsymbol{k}}^{\dagger} d_{\boldsymbol{Q}} c_{\beta,\boldsymbol{k}-\boldsymbol{Q}_{||}} + h. a.$$
(S5)

Here, $a_k^{\dagger}(a_k)$ denotes creation (annihilation) of a photon with in-plane wave number **k**, $c_{\alpha,k}^{\dagger}(c_{\alpha,k})$ of an electron with wave number **k** in subband α , and $d_q^{\dagger}(d_q)$ of a phonon with 3D wave vector **Q**. Furthermore, $\hbar\omega_{cav,k}$ is the energy of the bare cavity, $\hbar\omega_{LO}$ is the LO phonon energy, and $E_{a,k}$ is the energy of an electron in subband a with wave number **k**. The unscreened Coulomb and Fröhlich coupling matrix elements are given by:

$$\check{V}_{q}^{\alpha,\beta,\gamma,\delta} = \frac{e^{2}}{2\varepsilon_{s}A} \frac{1}{|q|} \int dz \,\psi_{\alpha}(z) \,\psi_{\gamma}(z) \int dz' \,\psi_{\beta}(z') \,\psi_{\delta}(z') \,e^{-|q||z-z'|},$$
(S6)
$$g_{Q}^{\alpha,\beta} = -i \,\sqrt{\frac{e^{2}\hbar\omega_{L0}}{2V} \left(\frac{1}{\varepsilon_{\infty}} - \frac{1}{\varepsilon_{s}}\right)} \frac{1}{|Q|} \int dz \,\psi_{\alpha}(z) \,\psi_{\beta}(z) \,e^{i \,Q_{\perp} z}.$$
(S7)

V=A L is the normalization volume and $\psi_{\alpha}(z)$ the wavefunction of the electronic states. The eigenstates of the confined electrons (i.e. wavefunctions $\psi_{\alpha}(z)$ and eigenenergies $E_{a,k}$) are calculated following the concept developed by Winkler et al based on the Fourier solution of the

Schroedinger equation using an eight band **k.p** Hamiltonian that fully accounts for non-parabolicity effects.

The screened Coulomb matrix elements are obtained from the unscreened ones by solving the tensor equation

$$V_{q}^{\alpha,\beta,\gamma,\delta} = \check{V}_{q}^{\alpha,\beta,\gamma,\delta} + \sum_{x,y} \check{V}_{q}^{\alpha,x,\gamma,y} \Pi_{q}^{x,y} V_{q}^{x,\beta,y,\delta}$$
(S8)

where $\Pi_q^{x,y}$ denotes the polarization function.

Supplementary Note 2: Absorption of the bare intersubband transition

Using

$$a(\omega) \sim \omega \,\Im(\frac{P(\omega)}{E(\omega)})$$
 (S9)

the absorption of the bare IST can be obtained directly from the Fourier-transformed microscopic polarization P(t)

$$P(t) = \sum_{k} d_{12} \, p_{k}^{12}(t) \,. \tag{S10}$$

We here determine the intersubband coherence p_k^{12} by solving the Heisenberg Equation in second-order Born-Markov approximation

$$\frac{d}{dt}p_{k}^{12} = \frac{i}{\hbar} < \left[H, c_{1,k}^{\dagger} c_{2,k}\right] > = \frac{d}{dt}p_{k}^{12}|_{0,cl} + \frac{d}{dt}p_{k}^{12}|_{HF} + \frac{d}{dt}p_{k}^{12}|_{scatt}.$$
(S11)

The free-carrier and carrier-field contributions are given by

$$\frac{d}{dt}p_{k}^{12}|_{0,cl} = \frac{i}{\hbar} \left(E_{1,k} - E_{2,k} \right) p_{k}^{12} + \frac{i}{\hbar} \int dz \ E(t) \ d_{12} \left(n_{1,k} - n_{2,k} \right), \tag{S12}$$

the first-order or Hartree-Fock contributions (exchange, exitonic, and depolarization effect) are given by

$$\frac{d}{dt} p_{k}^{12}|_{HF} = \frac{i}{\hbar} p_{k}^{12} \sum_{q \neq 0} \left(V_{\mathbf{q}}^{2112} - V_{\mathbf{q}}^{1111} \right) n_{1,\mathbf{k}-\mathbf{q}} - \frac{i}{\hbar} p_{k}^{12} \sum_{q \neq 0} \left(V_{\mathbf{q}}^{2112} - V_{\mathbf{q}}^{2222} \right) n_{2,\mathbf{k}-\mathbf{q}} \qquad (S13)$$

$$+ \frac{i}{\hbar} \left(n_{1,\mathbf{k}} - n_{2,\mathbf{k}} \right) \sum_{q \neq 0} V_{\mathbf{q}}^{1212} p_{\mathbf{k}-\mathbf{q}}^{12} - 2 \frac{i}{\hbar} V_{\mathbf{0}}^{2112} (n_{1,\mathbf{k}})$$

$$- n_{2,\mathbf{k}} \sum_{q \neq 0} p_{\mathbf{q}}^{12},$$

and the second-order or scattering contributions are given by

$$\frac{d}{dt}p_{k}^{12}|_{scatt} = \sum_{i=1,2} \left[-\frac{\pi}{\hbar} \Gamma_{d}^{i} p_{k}^{12} + \frac{\pi}{\hbar} \sum_{\mathbf{Q}_{\parallel}} \Gamma_{nd1}^{i} p_{k+\mathbf{Q}_{\parallel}}^{12} + \frac{\pi}{\hbar} \sum_{\mathbf{k}',q} \Gamma_{nd2}^{i} p_{\mathbf{k}'-\mathbf{q}}^{12} + \frac{\pi}{\hbar} \sum_{\mathbf{q}} \Gamma_{nd3}^{i} p_{\mathbf{k}+\mathbf{q}}^{12} - \frac{\pi}{\hbar} \sum_{\mathbf{k}'} \Gamma_{nd4}^{i} p_{\mathbf{k}'}^{12} \right].$$
(S14)

 $n_{i,\mathbf{k}}$ denotes the electron distribution functions for an electron in subband *i*. While the Hartree-Fock contributions renormalize the free-carrier and carrier-field contributions, the scattering contributions yield a de-phasing of the intersubband coherence $p_{\mathbf{k}}^{12}$. They consist of diagonal terms which depend on $p_{\mathbf{k}}^{12}$ at wave number **k**, and non-diagonal terms which depend on the intersubband coherence at different momenta **k**'', and thus couple different k states. Whereas the diagonal scattering contributions can be identified with the inverse of the T_2 time know from two-level atomic systems, the non-diagonal contributions yield a momentum dependent dephasing that strongly compensates the diagonal parts.

The diagonal scattering and non-diagonal contributions due to carrier-(LO) phonon and carriercarrier scattering read

$$\begin{split} \Gamma_{d,cp}^{1} &= \sum_{\mathbf{Q}} \left(\delta \left(-E_{1,k} + E_{1,k+\mathbf{Q}_{\parallel}} - \hbar \omega_{LO} \right) \left| g_{\mathbf{Q}}^{1,1} \right|^{2} \left[n_{\mathbf{Q}} \left(1 - n_{1,k+\mathbf{Q}_{\parallel}} \right) + \left(n_{\mathbf{Q}} \right) \right] \\ &+ 1 n_{1,k+\mathbf{Q}_{\parallel}} \right] \\ &+ \left(\delta \left(-E_{1,k} + E_{1,k+\mathbf{Q}_{\parallel}} + \hbar \omega_{LO} \right) \left| g_{\mathbf{Q}}^{1,1} \right|^{2} \left[(n_{\mathbf{Q}} + 1)(1 - n_{1,k+\mathbf{Q}_{\parallel}}) \right] \\ &+ n_{\mathbf{Q}} n_{1,k+\mathbf{Q}_{\parallel}} \right] \\ &+ \left(\delta \left(-E_{2,k} + E_{1,k+\mathbf{Q}_{\parallel}} - \hbar \omega_{LO} \right) \left| g_{\mathbf{Q}}^{1,2} \right|^{2} \left[n_{\mathbf{Q}} \left(1 - n_{1,k+\mathbf{Q}_{\parallel}} \right) + \left(n_{\mathbf{Q}} \right) \right] \\ &+ 1 n_{1,k+\mathbf{Q}_{\parallel}} \right] \\ &+ \left(\delta \left(-E_{2,k} + E_{1,k+\mathbf{Q}_{\parallel}} + \hbar \omega_{LO} \right) \left| g_{\mathbf{Q}}^{1,2} \right|^{2} \left[(n_{\mathbf{Q}} + 1)(1 - n_{1,k+\mathbf{Q}_{\parallel}}) + \left(n_{\mathbf{Q}} \right) \right] \\ &+ n_{\mathbf{Q}} n_{1,k+\mathbf{Q}_{\parallel}} \right] \end{split}$$

$$\begin{split} \Gamma_{d,cc}^{1} &= \sum_{\mathbf{k}',\mathbf{q}} (\delta(E_{1,\mathbf{k}} + E_{1,\mathbf{k}'} - E_{1,\mathbf{k}'-q} + E_{1,\mathbf{k}+q}) V_{q}^{1111} \left(2V_{q}^{1111}\right) \\ &\quad - V_{\mathbf{k}'-\mathbf{k}-q}^{1111} \left[n_{1,\mathbf{k}'} (1 - n_{1,\mathbf{k}'-q}) (1 - n_{1,\mathbf{k}+q}) \right. \\ &\quad + n_{1,\mathbf{k}'-q} n_{1,\mathbf{k}+q} (1 - n_{1,\mathbf{k}'}) \right] \\ &\quad + \delta(E_{1,\mathbf{k}} + E_{1,\mathbf{k}'} - E_{2,\mathbf{k}'-q} + E_{2,\mathbf{k}+q}) V_{q}^{1122} (2V_{q}^{1122} \\ &\quad - V_{\mathbf{k}'-\mathbf{k}-q}^{1122}) [n_{1,\mathbf{k}'} (1 - n_{2,\mathbf{k}'-q}) (1 - n_{2,\mathbf{k}+q}) \\ &\quad + n_{2,\mathbf{k}'-q} n_{2,\mathbf{k}+q} (1 - n_{1,\mathbf{k}'}) \right] \\ &\quad + \delta(E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{1,\mathbf{k}'-q} + E_{2,\mathbf{k}+q}) V_{q}^{1221} (2V_{q}^{1221} \\ &\quad - V_{\mathbf{k}'-\mathbf{k}-q}^{1212}) [n_{2,\mathbf{k}'} (1 - n_{1,\mathbf{k}'-q}) (1 - n_{2,\mathbf{k}+q}) \\ &\quad + n_{1,\mathbf{k}'-q} n_{2,\mathbf{k}+q} (1 - n_{2,\mathbf{k}'}) \right] \\ &\quad + \delta(E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{2,\mathbf{k}'-q} + E_{1,\mathbf{k}+q}) V_{q}^{1212} (2V_{q}^{1212} \\ &\quad - V_{\mathbf{k}'-\mathbf{k}-q}^{1212}) [n_{2,\mathbf{k}'} (1 - n_{2,\mathbf{k}'-q}) (1 - n_{2,\mathbf{k}+q}) \\ &\quad + n_{1,\mathbf{k}'-q} n_{2,\mathbf{k}+q} (1 - n_{2,\mathbf{k}'}) \right] \end{split}$$

$$\Gamma_{nd1}^{1} = \delta \left(-E_{1,k} + E_{1,k+Q_{\parallel}} - \hbar \omega_{LO} \right) g_{Q}^{1,1*} g_{Q}^{2,2} \left[n_{Q} \left(1 - n_{1,k+Q_{\parallel}} \right) + (n_{Q} + 1) n_{1,k+Q_{\parallel}} \right]$$

$$+ \left(\delta \left(-E_{1,k} + E_{1,k+Q_{\parallel}} + \hbar \omega_{LO} \right) g_{Q}^{1,1*} g_{Q}^{2,2} \left[(n_{Q} + 1)(1 - n_{1,k+Q_{\parallel}}) + n_{Q} n_{1,k+Q_{\parallel}} \right]$$

$$+ n_{Q} n_{1,k+Q_{\parallel}} \right]$$
(S17)

$$\Gamma_{nd2}^{1} = \sum_{\mathbf{k}',\mathbf{q}} (\delta (E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{1,\mathbf{k}'-\mathbf{q}} + E_{2,\mathbf{k}+\mathbf{q}}) V_{\mathbf{q}}^{2222} (2V_{\mathbf{q}}^{1221}$$

$$- V_{\mathbf{k}'-\mathbf{k}-\mathbf{q}}^{1212}) [n_{2,\mathbf{k}+\mathbf{q}} (1 - n_{1,\mathbf{k}}) (1 - n_{2,\mathbf{k}'}) + n_{1,\mathbf{k}} n_{2,\mathbf{k}'} (1 - n_{2,\mathbf{k}+\mathbf{q}})]$$

$$+ \delta (E_{1,\mathbf{k}} + E_{1,\mathbf{k}'} - E_{1,\mathbf{k}'-\mathbf{q}} + E_{1,\mathbf{k}+\mathbf{q}}) V_{\mathbf{q}}^{1221} (2V_{\mathbf{q}}^{1111}$$

$$- V_{\mathbf{k}'-\mathbf{k}-\mathbf{q}}^{1111}) [n_{1,\mathbf{k}+\mathbf{q}} (1 - n_{1,\mathbf{k}}) (1 - n_{1,\mathbf{k}'}) + n_{1,\mathbf{k}} n_{1,\mathbf{k}'} (1 - n_{1,\mathbf{k}+\mathbf{q}})])$$
(S18)

$$\Gamma_{nd3}^{1} = \sum_{\mathbf{k}',\mathbf{q}} (\delta(E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{2,\mathbf{k}'-\mathbf{q}} + E_{1,\mathbf{k}+\mathbf{q}}) V_{\mathbf{q}}^{2222} (2V_{\mathbf{q}}^{1212}$$

$$-V_{\mathbf{k}'-\mathbf{k}-\mathbf{q}}^{1221}) [n_{2,\mathbf{k}'-\mathbf{q}} (1 - n_{1,\mathbf{k}}) (1 - n_{2,\mathbf{k}'}) + n_{1,\mathbf{k}} n_{2,\mathbf{k}'} (1 - n_{2,\mathbf{k}'-\mathbf{q}})]$$

$$+ \delta(E_{1,\mathbf{k}} + E_{1,\mathbf{k}'} - E_{1,\mathbf{k}'-\mathbf{q}} + E_{1,\mathbf{k}+\mathbf{q}}) V_{\mathbf{q}}^{2121} (2V_{\mathbf{q}}^{1111} - V_{\mathbf{k}'-\mathbf{k}-\mathbf{q}}^{1111})$$

$$- V_{\mathbf{k}'-\mathbf{k}-\mathbf{q}}^{1111} [n_{1,\mathbf{k}'-\mathbf{q}} (1 - n_{1,\mathbf{k}}) (1 - n_{1,\mathbf{k}'}) + n_{1,\mathbf{k}} n_{1,\mathbf{k}'} (1 - n_{1,\mathbf{k}'-\mathbf{q}})])$$
(S19)

$$\Gamma_{nd4}^{1} = \sum_{\mathbf{k}',\mathbf{q}} (\delta(E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{2,\mathbf{k}'-q} + E_{1,\mathbf{k}+q}) V_{q}^{1221} (2V_{q}^{1212}$$

$$(S20)$$

$$- V_{\mathbf{k}'-\mathbf{k}-q}^{1221} [n_{1,\mathbf{k}} (1 - n_{2,\mathbf{k}'-q}) (1 - n_{1,\mathbf{k}+q}) \\
+ n_{2,\mathbf{k}'-q} n_{1,\mathbf{k}+q} (1 - n_{1,\mathbf{k}})] \\
+ \delta(E_{1,\mathbf{k}} + E_{2,\mathbf{k}'} - E_{1,\mathbf{k}'-q} + E_{2,\mathbf{k}+q}) V_{q}^{2121} (2V_{q}^{1221} \\
- V_{\mathbf{k}'-\mathbf{k}-q}^{1212} [n_{1,\mathbf{k}} (1 - n_{1,\mathbf{k}'-q}) (1 - n_{2,\mathbf{k}+q}) \\
+ n_{1,\mathbf{k}'-q} n_{1,\mathbf{k}+q} (1 - n_{1,\mathbf{k}})])$$

The contributions Γ_d^2 and Γ_{nd}^2 are obtained by exchanging the subband indices 1 and 2 in the expressions given for Γ_d^1 and Γ_{nd}^1 .

As an example we show in Supplementary Figure S1 the calculated absorption of sample B. To illustrate the impact of the different scattering contributions we also show the calculated absorption including only carrier-phonon scattering (cp), only carrier-carrier scattering (cc) and including the diagonal scattering contributions only (cp+cc, diag). As can be seen neglecting the non-diagonal contributions yields a dramatic overestimation of the line width. Comparing the line broadening due to the different scattering contributions (full, cc, cp) shows that carrier-carrier and carrier-phonon interactions are non-additive de-phasing mechanisms, due to interference effects of the diagonal and non-diagonal scattering contributions.

Supplementary Note 3: Polariton picture

The theory described here so far allows us to analyse our experiments for all values of excitation fields. This is achieved by developing our Hamiltonian in the electronic picture which captures more of the intersubband dynamics including the electron interaction with phonons and other electrons. In contrast to previous publications^{7-9,12,25} we do not assume a quasi-bosonic nature of the IST right from the beginning. This is only valid for weak excitation fields where a small number of electrons in the ground state are excited by the incoming light field and contribute to the light-matter interaction. Strong excitation fields violate this assumption and require the fermionic nature of the electrons to be included.

Nevertheless, it should be stressed here that our theory can be used to recover the published results for weak incoming field. Assuming that the rate of energy exchange between the cavity field and the intersubband transitions (IST) becomes much faster than the decay and the decoherence rates of both the cavity photons and the ISTs, polaritons are the elementary excitations (Bogoliubov-Hopfield transformation).

Including only collective excitations in the Hamiltonian and introducing IST operators, $b_k^{\dagger} = \frac{1}{\sqrt{N}}$ $\sum_q c_{2,k+q}^{\dagger} c_{1,q}$, the Hamiltonian of the system is reduced to

$$H \approx \sum_{k} \hbar \omega_{cav,k} \left(a_{k}^{\dagger} a_{k} + \frac{1}{2} \right) + \sum_{k} (E_{2,k} - E_{1,k}) b_{k}^{\dagger} b_{k}$$
(S21)

+ i
$$\hbar \Omega_R \sum_{k} (a_{-k} + a_k^{\dagger}) (b_k - b_{-k}^{\dagger}) + \frac{\hbar \Omega_R^2}{\omega_{21}} \sum_{k} (a_{-k} + a_k^{\dagger}) (a_k + a_{-k}^{\dagger})$$

+ $\sum_{k} V_0^{1221} (b_{-k} + b_k^{\dagger}) (b_k + b_{-k}^{\dagger}),$

or differently

$$H \approx \sum_{k} \hbar \omega_{cav,k} \left(a_{k}^{\dagger} a_{k} + \frac{1}{2} \right) + \sum_{k} \hbar \widetilde{\omega}_{21} B_{k}^{\dagger} B_{k}$$

$$+ i \hbar \widetilde{\Omega_{R}} \sum_{k} \left(a_{-k} + a_{k}^{\dagger} \right) \left(B_{k} - B_{-k}^{\dagger} \right) + \frac{\hbar \widetilde{\Omega_{R}^{2}}}{\omega_{21}} \sum_{k} \left(a_{-k} + a_{k}^{\dagger} \right) \left(a_{k} + a_{-k}^{\dagger} \right)$$
with $B_{k} = \frac{\widetilde{\omega}_{21} + \omega_{21}}{2\sqrt{\widetilde{\omega}_{21}\omega_{21}}} b_{k} + \frac{\widetilde{\omega}_{21} - \omega_{21}}{2\sqrt{\widetilde{\omega}_{21}\omega_{21}}} b_{k}^{\dagger}, \quad \widetilde{\omega}_{21} = \sqrt{\omega_{12}^{2} + \omega_{p}^{2}} \text{ and } \widetilde{\omega}_{21} = \widetilde{\omega}_{12}.$
(S22)

Above, ω_{12} is transition frequency of the IST without depolarization shift and ω_p equivalent to a quantum well (QW) plasma frequency. Both quantities are real valued and defined as

$$\omega_p^2 = f_{12} \frac{n_s e^2}{\varepsilon \, m^* L_{eff}} \tag{S23}$$

with

$$L_{eff} = \frac{\hbar f_{12}}{2m^* S E_{21}}, \qquad S = \int_{-\infty}^{\infty} dz \left(\int_{-\infty}^{z} dz' \ \psi_2(z') \ \psi_1(z') \right)^2, \tag{S24}$$

where n_s is the sheet carrier density inside the QW, m^* the effective electron mass, f_{12} the oscillator strength and $\psi_{1,2}$ the wavefunctions of the two electronic states.

Introducing polariton operators, $P_k = x_k a_k + y_k a_k^{\dagger} + z_k B_k + t_k B_k^{\dagger}$, the Hamiltonian is then obtained in diagonalized form which gives the energies of the upper and lower polaritons as the eigenenergies of the diagonalized Hamiltonian in the polariton picture (standard exciton-polariton theory). We end up with the solution for the on-resonance case:

$$\omega_{LP(UP)} = \sqrt{\widetilde{\omega}_{12}^2 + \Omega_R^2} \mp \Omega_R \tag{S25}$$

with $\Omega_R = \sqrt{f_W} \omega_p/2$ being the Rabi frequency. This result deviates from the conventional calculation of the polariton branches without the anti-resonant terms in the interaction Hamiltonian. However, it is interesting to note that the difference between the upper and lower polariton remains twice the Rabi frequency, recovering the conventional result.