

Seminar on the Optical Properties of Semiconductors

Polaritons

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May, 30. 2011

Wannier Equation

Wannier Equation including center-of-mass motion

$$-\left[\frac{\hbar^2 \nabla_{\mathbf{R}}^2}{2M} + \frac{\hbar^2 \nabla_{\mathbf{r}}^2}{2m_r} + V(r)\right] \psi(\mathbf{R}, \mathbf{r}) = E_{tot} \psi(\mathbf{R}, \mathbf{r}) \quad (1)$$

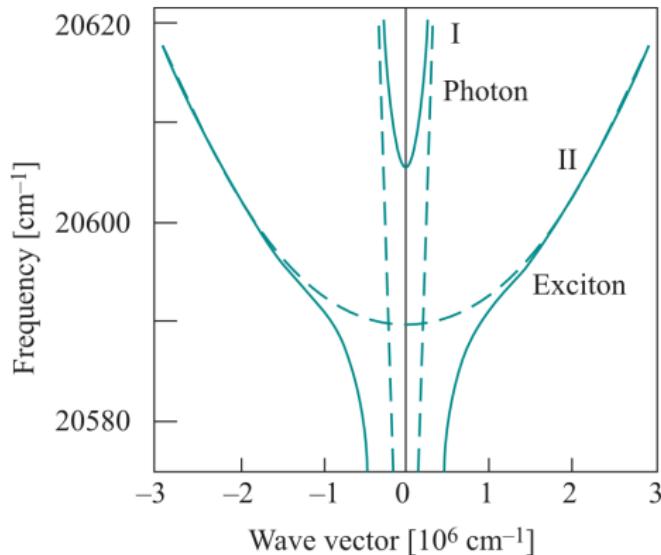
with total Mass $M = m_e + m_h$

$$\psi(\mathbf{R}, \mathbf{r}) = \frac{e^{i\mathbf{K}\cdot\mathbf{R}}}{L^{3/2}} \psi(\mathbf{r}) \quad (2)$$

plane wave for center-of-mass motion
and eigenvalues

$$E_{tot} = E_g + E_n + \frac{\hbar^2 K^2}{2M} \quad (3)$$

Exciton and Photon Dispersion



Photon dispersion

$$\omega = \frac{ck}{\sqrt{\epsilon}}$$

Exciton dispersion

$$\omega \propto k^2$$

Figure: Dispersion in CdS [Yu]

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Dielectric Theory

$$\begin{aligned}\epsilon(\omega) &= \epsilon_0 [1 + 4\pi\chi(\omega)] \\ &= \epsilon_0 \left[1 - 8\pi|d_{cv}|^2 \sum_n \frac{|\psi_n(\mathbf{r} = 0)|^2}{\hbar(\omega + i\delta) - E_g - E_n} \right]\end{aligned}\quad (4)$$

$$\left[-k^2 + \frac{\omega^2}{c^2} \epsilon(\omega) \right] \mathbf{E}_\omega e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} = 0 \quad (5)$$

with $\mathbf{E}_\omega \neq 0$

Transverse Eigenmodes

$$c^2 k^2 = \omega^2 \epsilon(\omega) \quad (6)$$

Polaritons without Spatial Dispersion

only 1s-excitons ($n = 0$)

$$\epsilon(\omega) \simeq \epsilon_0 \left[1 - 8\pi|d_{cv}|^2 \frac{|\psi_0(\mathbf{r} = 0)|^2}{\hbar(\omega + i\delta) - E_g + E_0} \right] \quad (7)$$

with $\hbar\Delta = 8\pi|d_{cv}|^2|\psi_0(\mathbf{r} = 0)|^2$ and $\hbar\omega_0 = E_g - E_0$

$$\epsilon(\omega) = \epsilon_0 \left(1 - \frac{\Delta}{\omega - \omega_0 + i\delta} \right) \quad (8)$$

using the Dirac identity $\lim_{\delta \rightarrow 0} \frac{1}{\omega - i\delta} = \mathcal{P} \frac{1}{\omega} + i\pi\delta(\omega)$
into eigenmode equation $c^2k^2 = \omega^2\epsilon(\omega)$

Polaritons without Spatial Dispersion

complex wave number $k = k' + ik''$

$$c^2 k^2 = \omega^2 \epsilon(\omega) \quad \text{in} \tag{9}$$

$$\text{real part} \quad \frac{\omega^2 \epsilon_0}{c^2} \left(1 - \frac{\Delta}{\omega - \omega_0} \right) = k'^2 - k''^2 \tag{10}$$

and

$$\text{imaginary part} \quad \pi \delta(\omega - \omega_0) \frac{\omega^2 \epsilon_0}{c^2} = 2k' k'' \tag{11}$$

for $\omega \neq \omega_0$ no absorption

Polaritons without Spatial Dispersion

Dispersion Relation

Polariton Dispersion

$$\omega \sqrt{\frac{\omega - \omega_0 - \Delta}{\omega - \omega_0}} = \frac{ck'}{\sqrt{\epsilon_0}} \quad (12)$$

with "band gap"

$$\omega_0 < \omega < \omega_0 + \Delta$$

$$\omega \simeq \frac{ck'}{\sqrt{\epsilon_0(1 + \Delta/\omega_0)}} \quad (13)$$

for $\omega \ll \omega_0$

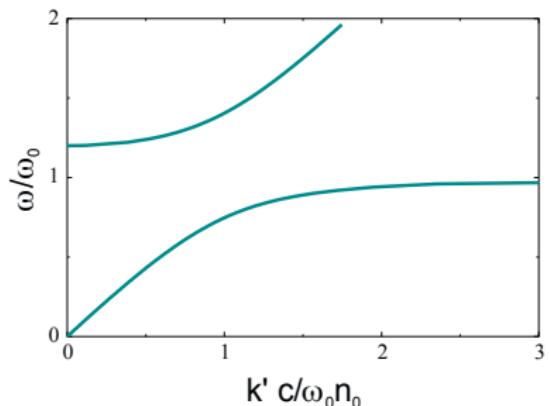


Figure: Polariton dispersion without spatial dispersion for $\Delta/\omega_0 = 0.2$ [Haug]

Polaritons with Spatial Dispersion and Damping

include photon momentum

$$\epsilon(\mathbf{K}, \omega) = \epsilon_0 \left(1 - \frac{\Delta}{\omega - \omega_0 - \frac{\hbar K^2}{2M} + i\gamma} \right) \quad (14)$$

eigenmode equation

$$\frac{c^2 K^2}{\epsilon_0} = \omega^2 \left(1 - \frac{\Delta}{\omega - \omega_0 - \frac{\hbar K^2}{2M} + i\gamma} \right) \quad (15)$$

Polaritons with Spatial Dispersion and Damping

Dispersion Relation

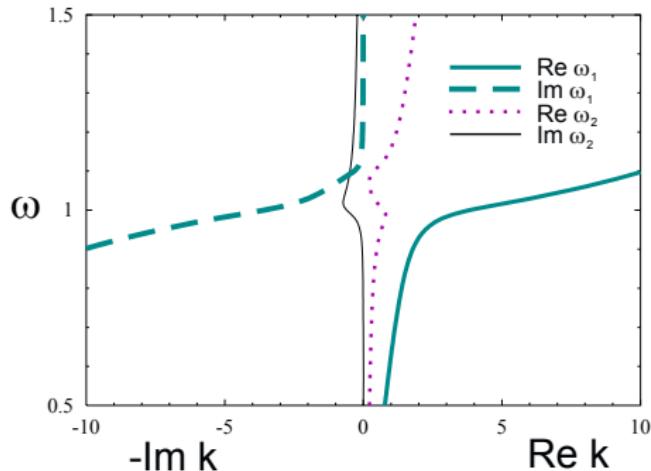


Figure: Polariton dispersion with spatial dispersion and damping for $\Delta/\omega_0 = 0.1$ [Haug]

Hamiltonian Theory of Polaritons

Preparations

hole operator $\beta_{-\mathbf{k}, -s}^\dagger \equiv a_{v, \mathbf{k}, s}$

el. operator $\alpha_{\mathbf{k}, s}^\dagger \equiv a_{c, \mathbf{k}, s}^\dagger$

el-hole pair operator

$$\alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}}^\dagger = |\mathbf{k}, -\mathbf{k}\rangle \langle 0|$$

exciton operator $B_{\nu, \mathbf{K}}^\dagger = |\nu \mathbf{K}\rangle \langle 0|$

$$\begin{aligned} B_{\nu, \mathbf{K}}^\dagger &= \sum_{\mathbf{k}, \mathbf{k}'} \langle \mathbf{k}, -\mathbf{k}' | \nu \mathbf{K} \rangle | \mathbf{k}, -\mathbf{k}' \rangle \langle 0 | \\ &= \sum_{\mathbf{k}, \mathbf{k}'} \langle \mathbf{k}, -\mathbf{k}' | \nu \mathbf{K} \rangle \alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}'}^\dagger \end{aligned}$$

$$\begin{aligned} \langle \mathbf{k}, -\mathbf{k}' | \nu \mathbf{K} \rangle &= \int d^3r d^3r' \langle \mathbf{k}, -\mathbf{k}' | \mathbf{r}, \mathbf{r}' \rangle \langle \mathbf{r}, \mathbf{r}' | \nu \mathbf{K} \rangle \\ &= \int d^3r d^3r' e^{-i\mathbf{k}\cdot\mathbf{r}} e^{i\mathbf{k}'\cdot\mathbf{r}'} e^{i\mathbf{K}\cdot(\mathbf{r}+\mathbf{r}')/2} \psi_\nu(\mathbf{r} - \mathbf{r}') \\ &= \delta[\mathbf{K} - (\mathbf{k} - \mathbf{k}')] \psi_\nu \left(\frac{\mathbf{k} + \mathbf{k}'}{2} \right) \end{aligned}$$

Hamiltonian Theory of Polaritons

$$B_{\nu, \mathbf{K}}^\dagger = \sum_{\mathbf{k}, \mathbf{k}'} \delta[\mathbf{K} - (\mathbf{k} - \mathbf{k}')] \psi_\nu \left(\frac{\mathbf{k} + \mathbf{k}'}{2} \right) \alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}}^\dagger \quad (16)$$

Exciton Creation Operator

$$B_{\nu, \mathbf{K}}^\dagger = \sum_{\mathbf{k}} \psi_\nu(\mathbf{k} - \mathbf{K}/2) \alpha_{\mathbf{k}}^\dagger \beta_{\mathbf{K}-\mathbf{k}}^\dagger \quad (17)$$

Hamiltonian Theory of Polaritons

Exciton Commutator

$$\begin{aligned}[B_{0,0}, B_{0,0}^\dagger] &= \sum_{\mathbf{k}, \mathbf{k}'} \psi_0(k) \psi_0^*(k') \left[\beta_{-\mathbf{k}} \alpha_{\mathbf{k}}, \alpha_{\mathbf{k}'}^\dagger \beta_{-\mathbf{k}'}^\dagger \right] \\ &= \sum_{\mathbf{k}} |\psi_0(k)|^2 (1 - \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} - \beta_{-\mathbf{k}}^\dagger \beta_{-\mathbf{k}})\end{aligned}\quad (18)$$

$$\langle [B_{0,0}, B_{0,0}^\dagger] \rangle = 1 - \mathcal{O}(na_0^3) \quad (19)$$

→ boson particle at low densities

Free Excitons

$$\mathcal{H}_0 = \sum_{\mathbf{k}} \hbar e_{\nu k} B_{\nu, \mathbf{k}}^\dagger B_{\nu, \mathbf{k}} \quad (20)$$

Hamiltonian Theory of Polaritons

Interaction Hamiltonian

with the wavenumber of the photon q

$$\begin{aligned}\mathcal{H}_I &= - \sum_{\mathbf{k}, \mathbf{q}} d_{cv} \left[\alpha_{\frac{1}{2}\mathbf{q}+\mathbf{k}}^\dagger \beta_{\frac{1}{2}\mathbf{q}-\mathbf{k}}^\dagger \mathcal{E}(\mathbf{q}) e^{-i\omega_q t} + h.c. \right] \\ &= - \sum_{\mathbf{k}, \mathbf{q}, \nu} d_{cv} \left[\psi_\nu^*(\mathbf{k}) B_{\nu, \mathbf{q}}^\dagger \mathcal{E}(\mathbf{q}) e^{-i\omega_q t} + h.c. \right]\end{aligned}\quad (21)$$

with

$$\begin{aligned}\sum_{\nu} \psi_\nu^*(\mathbf{k}) B_{\nu, \mathbf{q}}^\dagger &= \sum_{\nu \mathbf{k}'} \psi_\nu^*(\mathbf{k}) \psi_\nu(\mathbf{k}' - \mathbf{q}/2) \alpha_{\mathbf{k}'}^\dagger \beta_{\mathbf{q}-\mathbf{k}'}^\dagger \\ &= \sum_{\mathbf{k}'} \delta_{\mathbf{k}, \mathbf{k}' - \mathbf{q}/2} \alpha_{\mathbf{k}'}^\dagger \beta_{\mathbf{q}-\mathbf{k}'}^\dagger = \alpha_{\frac{1}{2}\mathbf{q}+\mathbf{k}}^\dagger \beta_{\frac{1}{2}\mathbf{q}-\mathbf{k}}^\dagger\end{aligned}\quad (22)$$

Field Quantization

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \sum_{\mathbf{k}, \lambda} \sqrt{\frac{2\pi c^2 \hbar}{\omega_k}} [b_{\lambda \mathbf{k}} \mathbf{u}_{\lambda \mathbf{k}}(\mathbf{r}) + b_{\lambda \mathbf{k}}^\dagger \mathbf{u}_{\lambda \mathbf{k}}^*(\mathbf{r})] \quad (23)$$

with field

$$\mathbf{u}_{\lambda \mathbf{k}} = \frac{e^{i \mathbf{k} \cdot \mathbf{r}}}{L^{3/2}} \mathbf{e}_{\lambda \mathbf{k}} \quad (24)$$

in wave equation

$$\left(\frac{\partial^2}{\partial t^2} - c^2 \nabla^2 \right) \hat{\mathbf{A}} = 0 \quad \rightarrow \quad \frac{\partial^2}{\partial t^2} b_{\lambda \mathbf{k}} = -\omega_k^2 b_{\lambda \mathbf{k}} \quad (25)$$

$$\mathcal{E}(\mathbf{q}, t) = -\frac{1}{c} \frac{\partial A(\mathbf{q}, t)}{\partial t} = i \sqrt{2\pi \hbar \omega_q} (b_{\mathbf{q}} - h.c.) \quad (26)$$

and $\omega_q = \frac{cq}{\sqrt{\epsilon_0}}$

Hamiltonian Theory of Polaritons

Including non resonant Terms

Interaction Hamiltonian

$$\mathcal{H}_I = i\hbar \sum_{\mathbf{q}} g_{\nu q} (B_{\mathbf{q}} + B_{-\mathbf{q}}^\dagger) (b_{-\mathbf{q}} - b_{\mathbf{q}}^\dagger) \quad (27)$$

$$\hbar g_{\nu q} = d_{cv} \psi_\nu^*(\mathbf{r} = 0) \sqrt{\pi \hbar \omega_q / 2} \quad (28)$$

diagonalized by Hopfield transformation

$$p_{i,\mathbf{q}} = u_{i,1,\mathbf{q}} B_{\mathbf{q}} + u_{i,2,\mathbf{q}} b_{\mathbf{q}} + u_{i,3,\mathbf{q}} B_{-\mathbf{q}}^\dagger + u_{i,4,\mathbf{q}} b_{-\mathbf{q}}^\dagger \quad (29)$$

Hamiltonian Theory of Polaritons

Interaction Hamiltonian II

only resonant terms

$$\mathcal{H}_I = -i\hbar \sum_{\mathbf{q}, \nu} g_{\nu\mathbf{q}} (B_{\nu\mathbf{q}}^\dagger b_{\mathbf{q}} - h.c.) \quad (30)$$

Exciton-Photon Hamiltonian

$$\mathcal{H}_{xp} = \hbar \sum_{\mathbf{q}} \left[\sum_{\nu} e_{\nu\mathbf{q}} B_{\nu\mathbf{q}}^\dagger B_{\nu\mathbf{q}} + \omega_q b_{\mathbf{q}}^\dagger b_{\mathbf{q}} - i \sum_{\nu} g_{\nu\mathbf{q}} (B_{\nu\mathbf{q}}^\dagger b_{\mathbf{q}} - h.c.) \right] \quad (31)$$

Hamiltonian Theory of Polaritons

Diagonalization

Bogoliubov transformation

$$p_{\mathbf{q}} = u_{\mathbf{q}} B_{\mathbf{q}} + v_{\mathbf{q}} b_{\mathbf{q}} \quad (32)$$

$$[p_{\mathbf{q}}, p_{\mathbf{q}}^\dagger] = |u_{\mathbf{q}}|^2 + |v_{\mathbf{q}}|^2 = 1 \quad (33)$$

Polariton Hamiltonian

$$\mathcal{H}_p = \hbar \sum_{\mathbf{q}} \Omega_q p_{\mathbf{q}}^\dagger p_{\mathbf{q}} \quad (34)$$

Hamiltonian Theory of Polaritons

Dispersion Relation

evaluate the commutator $[p, H_p]/\hbar = [p, H_{xp}]/\hbar$

$$\begin{aligned}\Omega_q p_q &= \Omega_q(u_q B_q + v_q b_q) \\ &= u_q(e_q B_q - ig_q b_q) + v_q(\omega_b q + ig_q B_q)\end{aligned}\quad (35)$$

B_q and b_q have to be zero independently

$$0 = (\Omega_q - e_q)u_q + ig_q v_q \quad (36)$$

$$0 = -ig_q u_q + (\Omega_q - \omega_q)v_q \quad (37)$$

solution of linear system of equations with $\det = 0$

Polariton Dispersion

$$\Omega_{q,1,2} = \frac{1}{2}(e_q + \omega_q) \pm \frac{1}{2}\sqrt{(e_q - \omega_q)^2 + 4g_q^2} \quad (38)$$

Hamiltonian Theory of Polaritons

Dispersion Relation

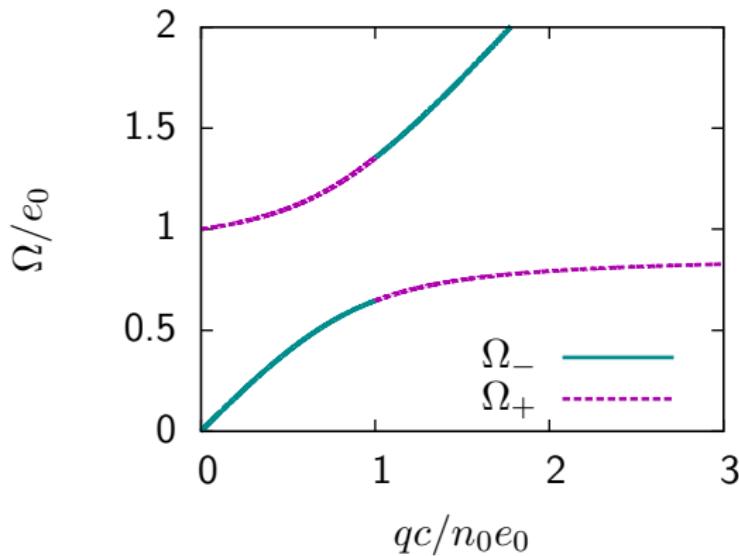


Figure: Polariton dispersion from Hamiltonian theory with $e_q = e_0$, $4g_q^2 = \Delta \cdot \omega_q$ and $\Delta/e_0 = 0.2$

Hamiltonian Theory of Polaritons

Photon and Exciton Contributions

$$p_{\mathbf{q}} = u_{\mathbf{q}} B_{\mathbf{q}} + v_{\mathbf{q}} b_{\mathbf{q}}$$

$$u_{\mathbf{q},1} = \sqrt{\frac{\Omega_{\mathbf{q},1} - \omega_{\mathbf{q}}}{2\Omega_{\mathbf{q},1} - e_{\mathbf{q}} - \omega_{\mathbf{q}}}}$$

and

$$v_{\mathbf{q},1} = i \sqrt{\frac{\Omega_{\mathbf{q},1} - e_{\mathbf{q}}}{2\Omega_{\mathbf{q},1} - e_{\mathbf{q}} - \omega_{\mathbf{q}}}}$$

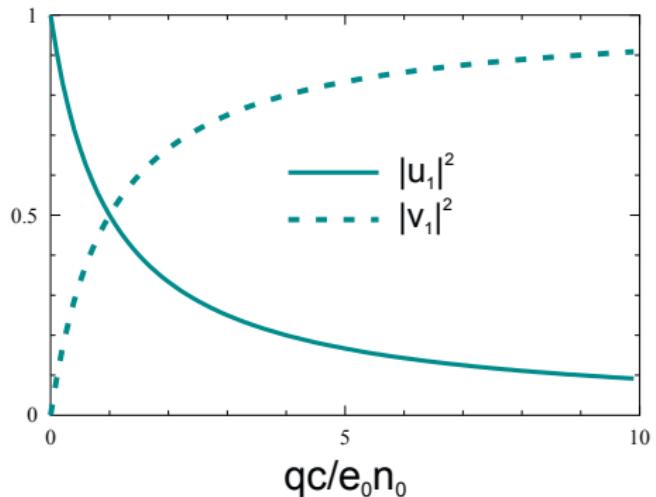
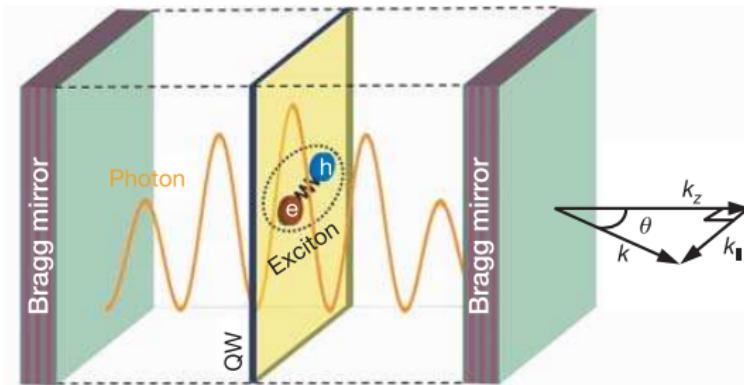


Figure: Dispersion of $|u_i|^2$ and $|v_i|^2$ for the upper polariton branch[Haug]

Microcavity Polaritons



with
eigenfrequencies

$$\omega_{0,k_{\parallel}} = \frac{c}{n} \sqrt{\frac{\pi^2}{L^2} + k_{\parallel}^2}$$

Figure: Microcavity with 2D Quantum well inside
[Kasprzak]

$$L = \frac{\lambda}{2} \quad \rightarrow \quad \mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}_0 \sin\left(\frac{\pi z}{L}\right) e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}} \quad (39)$$

Microcavity Polaritons

$$\mathcal{H} = \hbar \sum_{\mathbf{k}_{\parallel}} \left[e_{0,k_{\parallel}} B_{0,\mathbf{k}_{\parallel}}^{\dagger} B_{0,\mathbf{k}_{\parallel}} + \omega_{0,k_{\parallel}} b_{0,\mathbf{k}_{\parallel}}^{\dagger} b_{0,\mathbf{k}_{\parallel}} - i g_{k_{\parallel}} (B_{0,\mathbf{k}_{\parallel}}^{\dagger} b_{0,\mathbf{k}_{\parallel}} - \text{h.c.}) \right] \quad (40)$$

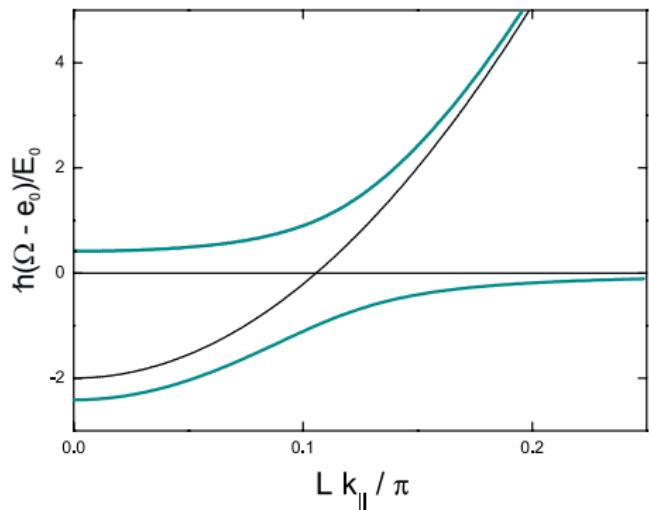
$$\hbar g_{k_{\parallel}} = d_{cv} \psi_0^{2D}(r=0) \sqrt{\pi \hbar \omega_{0,k_{\parallel}}} \quad (41)$$

Dispersion Relation inside Quantum Well

$$\Omega_{k_{\parallel},1,2} = \frac{1}{2} (e_{0,k_{\parallel}} + \omega_{0,k_{\parallel}}) \pm \frac{1}{2} \sqrt{(e_{0,k_{\parallel}} - \omega_{0,k_{\parallel}})^2 + 4g_{k_{\parallel}}^2} \quad (42)$$

Microcavity Polaritons

Dispersion Relation



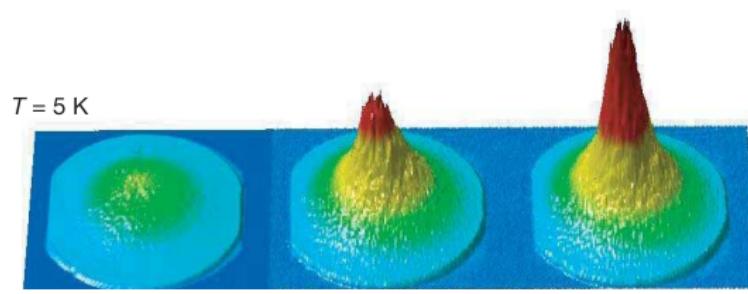
tunable coupling

$$g_{k_{\parallel}} \propto \sqrt{\omega_{0,k_{\parallel}}}$$
$$\omega_{0,k_{\parallel}} = \frac{c}{n} \sqrt{\frac{\pi^2}{L^2} + k_{\parallel}^2}$$

Figure: Microcavity Polariton spectrum [Haug]

Application I

Bose-Einstein Condensation of Exciton Polaritons



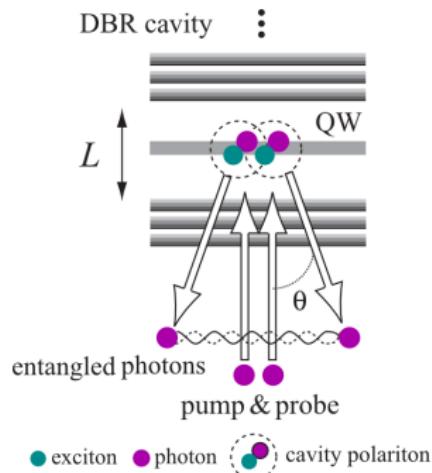
- ground state emission
- spontaneous polarization
- coherence length

Figure: Far-field polariton emission [Kasprzak]

But: interaction, thus no ideal Bose-gas

Application II

Generation of Entangled Photons



other applications:

- use long lifetime
- mediation of e.g. spin coupling

Figure: Entangled-photon generation from cavity bipolariton [Oka]

Summary

- Interaction between photons and excitons in semiconductor
- Coupling introduces joint dispersion with gab described by quasiparticles
- Dielectric function
- Microscopic theory
 - Properties of quasiparticles
 - Distinguish photon and exciton contributions
 - Microcavity polaritons
 - Left out damping (phonons)
- Applications
 - BEC in microcavities
 - Generation of entangled photons

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Longitudinal-transverse Splitting

Table: LT Splitting for 1s exciton-polaritons at $T \leq 4.2$ K

Material	$\hbar\Delta_{LT}$ / meV
GaAs	≈ 0.1
CdTe	≈ 0.4
ZnSe	≈ 1.3
CdS	$\approx 2(A)$
CuCl	$5.4 - 5.7(Z_3)$

$$\Delta_{LT} \propto \frac{|d_{vc}|^2}{a_0^3} \quad (43)$$

[Schäfer and Wegener. *Semiconductor optics and transport phenomena*. Springer 2002]