

INTERNATIONAL ATOMIC ENERGY AGENCY UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS

SMR.769 - 6

#### **WORKSHOP ON** "NON-LINEAR ELECTROMAGNETIC INTERACTIONS IN SEMICONDUCTORS"

1 - 10 AUGUST 1994

"Quantum theory of quantum well polaritons in semiconductor microcavities"

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# Quantum theory of quantum well polaritons in semiconductor microcavities

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# Exciton-polariton: coherent excitation of exciton state & electromagnetic field

Constituent equation

$$\frac{1}{\omega_0^2}\ddot{P} + \frac{\Gamma}{\omega_0^2}\dot{P} - \frac{\hbar}{M\omega_0}\nabla^2 P + P = \chi E$$

& Maxwell equations

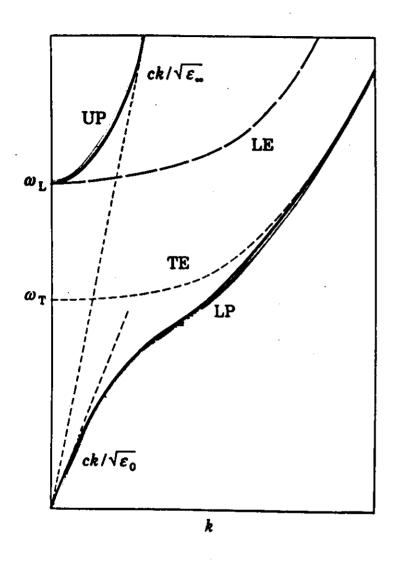
Dielectric function

$$\mathcal{E}(\Omega, \mathbf{k}) = \mathcal{E}_{\infty} + \frac{4\pi \chi \omega_0^2}{\omega_k^2 - \Omega^2 + i\Gamma\Omega}$$

$$\omega_{\mathbf{k}} = \omega_0 + \frac{\hbar}{2M} k^2$$

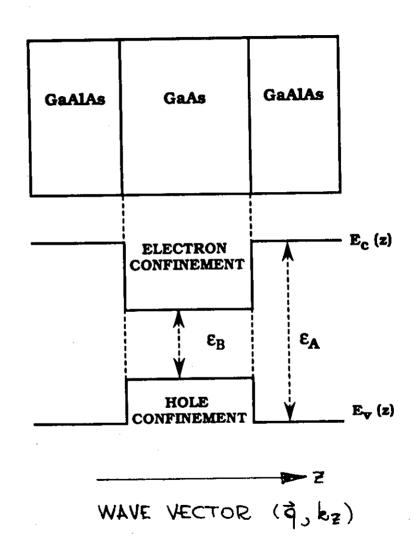
Longitudinal modes  $\varepsilon(\Omega, \mathbf{k}) = 0$ 

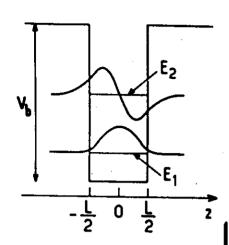
Transverse modes 
$$\varepsilon(\Omega, \mathbf{k}) = \frac{c^2 k^2}{\Omega^2}$$



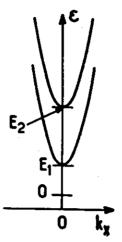
Exciton-polariton: coherent excitation of exciton state & electromagnetic field

## **QUANTUM WELL**



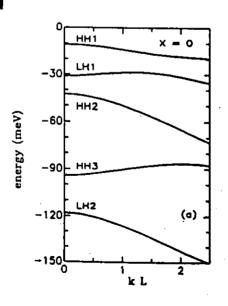


BOUND ELECTRONIC STATES IN A Q.W

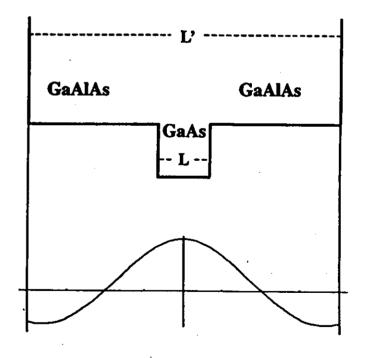


#### DISPERSION OF THE CONDUCTION SUBBANDS

DISPERSION OFTHE



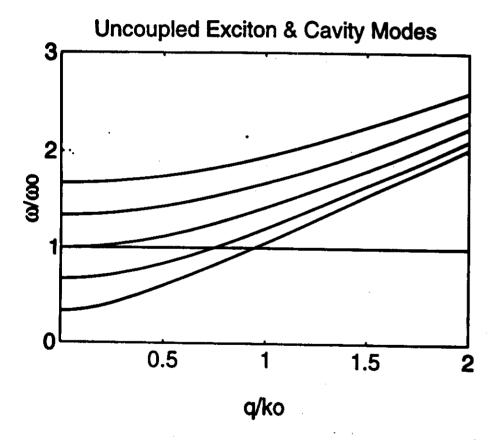
## **CLOSED CAVITY**



**OPTICAL FIELD** 

EXCITON : A-L BOILL

#### Finite cavity with perfect reflecting mirrors Finite cavity L' > QW thickness L



#### **HAMILTONIAN**

$$\begin{split} H = \sum_{\mathbf{q}} \hbar \omega_{\mathbf{q}} A_{\mathbf{q}}^{2^{+}} A_{\mathbf{q}}^{2} + \sum_{\mathbf{q}, k_{z}, \lambda} \hbar \mathbf{v} \, \sqrt{\mathbf{q}^{2} + k_{z}^{2}} \, A_{\mathbf{q}, k_{z}, \lambda}^{1^{+}} A_{\mathbf{q}, k_{z}, \lambda}^{1} \\ + i \sum_{\mathbf{q}, k_{z}, \lambda} C_{\mathbf{q}, k_{z}}^{\lambda} (A_{-\mathbf{q}}^{2} - A_{\mathbf{q}}^{2^{+}}) (A_{\mathbf{q}, k_{z}, \lambda}^{1} + A_{-\mathbf{q}, k_{z}, \lambda}^{1^{+}}) \\ + \left( \frac{1}{\hbar \omega_{\mathbf{q}}} \right) \sum_{\substack{\mathbf{q}, \mathbf{q}', \\ k_{z}, k_{z}', \lambda'}} C_{\mathbf{q}, k_{z}}^{\lambda' *} C_{\mathbf{q}', k_{z}'}^{\lambda' *} (A_{\mathbf{q}', k_{z}', \lambda'}^{1} + A_{-\mathbf{q}', k_{z}', \lambda'}^{1^{+}}) (A_{\mathbf{q}, k_{z}, \lambda}^{1} + A_{-\mathbf{q}, k_{z}, \lambda}^{1^{+}}) \end{split}$$

$$C_{\mathbf{q},k_{z}}^{\lambda} = \sqrt{\frac{4\pi\hbar v}{L'\sqrt{q^{2} + k_{z}^{2}}}} \omega_{q} \mu_{cv} \cdot \hat{\mathbf{e}}_{\mathbf{q},k_{z},\lambda} \frac{1}{c} F(0) \frac{1}{L} \int_{-L'/2}^{L'/2} dz \rho(z) e^{ik_{z}z} ,$$

$$\omega_q = \omega_0 + \gamma q^2$$
 : exciton frequency

$$v = c / \sqrt{\varepsilon_{\infty}}, \qquad k_0 = \omega_0 / v$$

 $\mu_{cv}$ : dipole matrix element

F(0): envelope function of the exciton at x = 0

 $\lambda$ : polarization index

 $\rho(z)$ : product of the confinement functions for e & h

 $A_{\mathbf{q},k_{\mathbf{r}},\lambda}^{1}$  and  $A_{\mathbf{q}}^{2}$  are Bose operators for photons & excitons

Only even TE cavity modes are considered

# TRANSFORMATION FROM FREE EXCITONS & PHOTONS

to

#### **UPPER & LOWER POLARITONS**

$$B_{\mathbf{q}}^{l} = \sum_{k_{z},\lambda} W_{l}(k_{z},\mathbf{q}) A_{\mathbf{q},k_{z},\lambda}^{1} + X_{l}(\mathbf{q}) A_{\mathbf{q}}^{2} + \sum_{k_{z},\lambda} Y_{l}(k_{z},\mathbf{q}) A_{-\mathbf{q},k_{z},\lambda}^{1^{+}} + Z_{l}(\mathbf{q}) A_{-\mathbf{q}}^{2^{+}}$$

*l* indicates the polariton modes

#### **Dispersion**

$$\Omega^{2} - \omega_{q}^{2} + \frac{4\omega_{q}}{\hbar^{2}} \frac{\Omega^{2}}{\omega_{q}^{2}} \sum_{k_{z},\lambda} \frac{v |Q|}{v^{2} |Q|^{2} - \Omega^{2}} |C_{q,k_{z}}^{\lambda}|^{2} = 0$$

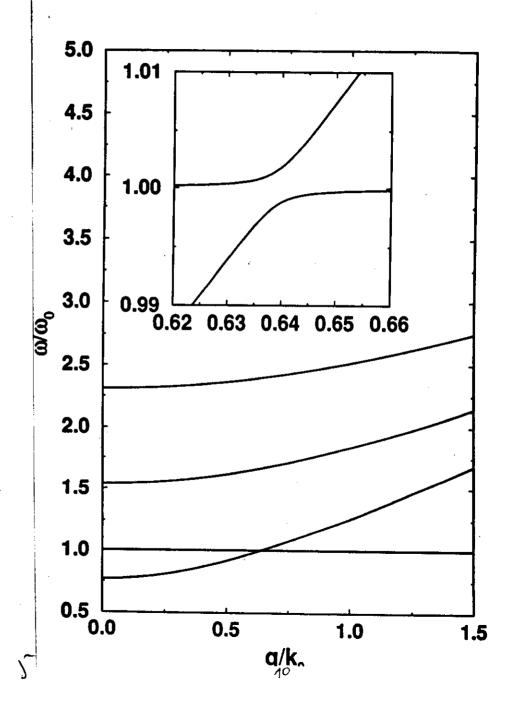
$$|Q| = \sqrt{k_{z}^{2} + q^{2}}$$

LOWER POLARITON  $\Omega < vq < v|Q|$ 

**UPPER POLARITON**  $\Omega \ge vq$ 

#### **POLARITON MODES IN A CLOSED CAVITY**

The anticrossing behaviour is shown in detail



The  $A^2$ -term is neglected. Cavity mode:  $k_z = 2\pi/L'$ Exciton frequency  $\omega_0$ ;  $k_0 \equiv \omega_0/v$ ;  $Q^2 \equiv (q^2 + k_z^2)/k_0^2$ Fixed polarization

$$C_{\mathbf{q},k_c}^{\lambda} = \sqrt{\frac{4\pi\hbar\nu}{L'|Q|}}\omega_{0}\mu_{cv}\cdot\hat{\mathbf{e}}_{\mathbf{q},k_c,\lambda}\frac{1}{c}F(0)\frac{1}{L}\int_{-L'/2}^{L'/2}dz\rho(z)e^{ik_zz}$$

$$\left| D_{\mathbf{q}}(k_z) \right|^2 \equiv \left| C_{\mathbf{q},k_z}^{\lambda} \right|^2 Q / \hbar^2 \omega_0^2$$

Approximate dispersion:

$$(1-\omega_l^2)(Q^2-\omega_l^2)=4|D(k_r)|^2$$
  $l=1, 2$ 

(dispersion for 2 - dimensional polaritons)

Polariton coefficients:

$$\left| X_{\omega_l}(\mathbf{q}) \right|^2 = \frac{(1+\omega_l)^2 (Q^2 - \omega_l^2)}{4\omega_l (1+Q^2 - 2\omega_l^2)}$$

For a resonant cavity ( $\omega \approx 1$ ) & normal incidence

$$\left|X_{\omega_l}(\mathbf{q})\right|^2 \to \frac{(1+\omega_l)^2}{8\omega_l} \approx \frac{1}{2}$$
 (exciton component)

$$\left|W_{\omega_i}(\mathbf{q}, k_z)\right|^2 \to \frac{(1+\omega_i)^2}{8\omega_i} \approx \frac{1}{2}$$
 (radiation component)

**VOLUME 69, NUMBER 23** 

PHYSICAL REVIEW LETTERS

7 DECEMBER 19

#### Observation of the Coupled Exciton-Photon Mode Splitting in a Semiconductor Quantum Microcavity

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(Received 12 May 1992)

The spectral response of a monolithic semiconductor quantum microcavity with quantum wells as the active medium displays mode splittings when the quantum wells and the optical cavity are in resonance. This effect can be seen as the Rabi vacuum-field splitting of the quantum-well excitons, or more classically as the normal-mode splitting of coupled oscillators, the excitons and the electromagnetic field of the microcavity. An exciton oscillator strength of  $4 \times 10^{12}$  cm<sup>-2</sup> is deduced for 76-Å quantum wells.

PACS numbers: 42.50.-p, 71.35.+z, 73.20.Dx, 78.45.+h

# Third International Conference on OPTICS OF EXCITONS IN CONFINED SYSTEMS MONTPELLIER, AUGUST 1993.

Room temperature exciton-photon Rabi splitting in a semiconductor microcavity.

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Strong exciton-photon coupling regime can be achieved in a semiconductor microcavity up to room temperature. In this regime, the Fabry-Pérot photon mode of the cavity and the exciton electronic mode are no longer eigenstates of the coupled system. Both states are strongly coupled and vacuum field Rabi splitting occurs. Analogies with atomic physics, conditions for strong coupling, experimental results and their implications are discussed.

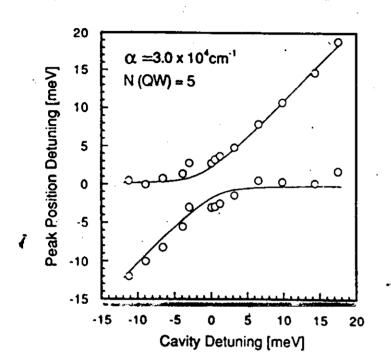
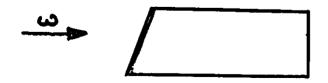


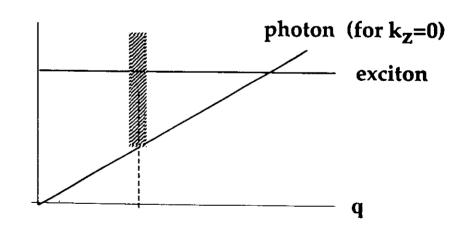
FIG. 3. Reflectivity peak positions as a function of cavity detuning for a five-quantum-well sample at T=5 K. The theoretical fit is obtained through a standard multiple-interference analysis of the DBR-Fabry-Pérot-quantum-well structure.



## INFINITE CAVITY – UPPER POLARITON $(L' \rightarrow \infty)$

For fixed q, the exciton frequency  $\omega_q$  is embedded in the photon continuum:  $vq \ge \omega_q \ge v\sqrt{k_z^2+q^2}$ . Polaritons appear as Fano resonances of frequency  $\Omega^{j(UP)}(q)$  and lifetime  $1/\Gamma^{j(UP)}(q)$ . The polariton spectrum  $\Omega^{j(UP)}(q)$  is continous in j and q.

#### frequency



**UPPER POLARITON – CLASSICAL RESONANCE**The mean polariton number

 $<\alpha({\bf q})_{\it exciton}, 0_{\it photon} \mid B_q^{l\,(UP)^+} B_q^{l\,(UP)} \mid \alpha({\bf q})_{\it exciton}, 0_{\it photon}>$  has its maximum value for the classical resonance frequency and a width coinciding with the classical lifetime broadening.

 $|\alpha(q)|_{exciton} > is a coherent exciton state of wave vector q.$ 

alternative approach

**UPPER POLARITON – CLASSICAL RESONANCE Finite Cavity L' >> QW thickness L.** 

 $\mathcal{D}_{j}(\Omega^{UP}) = 2\pi q \frac{\partial q}{\partial \Omega^{UP}}$  is the density of states of the upper polariton of the branch j.

The maximum of  $\mathcal{D}_j(\Omega^{UP})$  for each j coincides with the classical resonance frequency. The width of the extrema corresponds to the classical lifetime broadening.

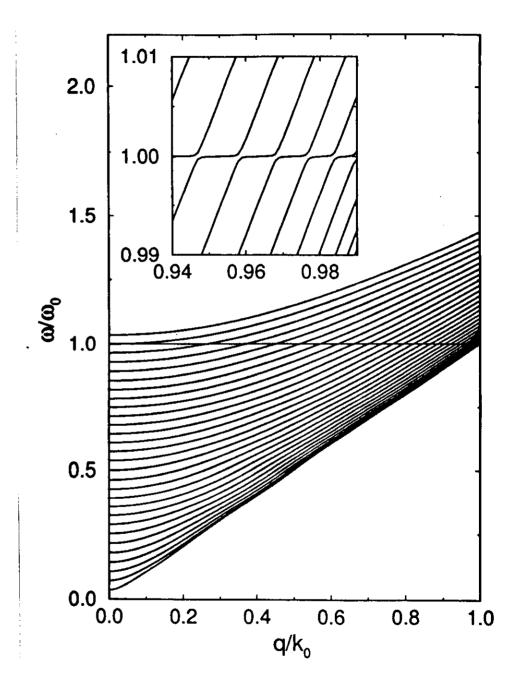
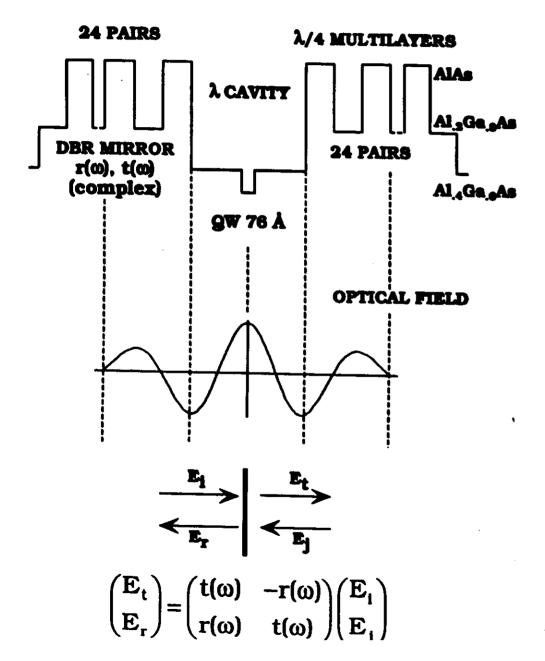


FIG. 3

### **CAVITY WITH LOSSES**



A semiconductor microcavity is a planar Fabry-Pérot whose mirrors are multilayer structures built with alternating layers of two different refraction indexes and the same thickness  $\lambda/4$ , where  $\lambda$  is the resonance wave-length of the Fabry-Pérot.

A structure of this kind, called distributed Bragg reflector (DBR), presents a very high reflectivity in a given frequency interval around the resonance.

The DBR reflection coefficient for the electric field has a phase equal to zero, whereas for metallic mirrors this phase is equal to  $\pi$  Standing waves inside the DBR present antinodes instead of nodes, as for a metallic mirror, at the cavity boundaries.

Boundary conditions:  $1+|\rho|\exp[i\phi] = \tau$ .

Metal mirror :  $\phi = \pi$ . DBR mirror :  $\phi = 0$ .

#### **QUASIMODE APPROACH**

In order to include the feature of an open cavity (R < 1), the quasimode approach is used. The cavity mode a is coupled to the continuum of the external radiation modes by the following Hamiltonian

$$H_{\rm int} = \int d\omega \left\{ V^*(\omega) \boldsymbol{a} [b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega)] + h.c. \right\}$$

The relation between  $V(\omega)$  and the features of the DBR's is obtained by comparing the quasimodes density of state with the solution of the Maxwell equations in the limit of high reflectivity:

$$|V(\omega)|^2 = |t(\omega)|^2 \sqrt{4\pi} L'$$

 $t(\omega)$  is the complex transission coefficient of the DBR.

For normal incidence (q=0),  $k_z = 2\pi/L'$ , and considering one cavity mode only (two polariton modes) the photon operator in terms of polaritons reads

$$\mathbf{a} = W_{k_{z}}(\omega_{1}) B^{1+} + W_{k_{z}}(\omega_{2}) B^{2+}$$
$$-Y_{k_{z}}^{*}(\omega_{1}) B^{1} - Y_{k_{z}}^{*}(\omega_{2}) B^{2}$$

## CAVITY WITH LOSSES – QUASIMODE SCHEME EMPTY CAVITY

RADIATION FIELD INSIDE & OUTSIDE
THE CAVITY ARE DISTINCT

INSIDE THE CAVITY: ONE FIELD MODE  $a^{\#}$ 

OUTSIDE THE CAVITY: FREE FIELD  $b_{\hat{k}}^{\#}(\omega), b_{-\hat{k}}^{\#}(\omega)$ 

The two fields are coupled through

$$H_{\rm int} = \int d\omega \left\{ V^{\bullet}(\omega) a^{+} [b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega)] + h.c. \right\}$$

k : unit vector in the growth direction

#### DIAGONALIZATION BY THE FANO METHOD

$$c(\omega) = \alpha(\omega) \ a + \int \mathrm{d}\omega' \left\{ \beta_{\omega'}(\omega) b_{\hat{\mathbf{k}}}(\omega') + \gamma_{\omega'}(\omega) b_{-\hat{\mathbf{k}}}(\omega') \right\}$$

$$\alpha(\omega) = \frac{\sqrt{2}V^*(\omega)}{2\pi|V(\omega)|^2 + i(\omega - \omega_0 - F(\omega))}$$

 $|\alpha(\omega)|^2$ : Lorentzian line shape

$$F(\omega) = \wp \int d\omega' \frac{2|V(\omega)|^2}{\omega - \omega'}$$

#### **OPEN CAVITY**

#### **EXACT VERSUS QUASIMODE SCHEME**

- (1) Exact scheme: Solution of the Mawell equations with the appropriate boundary conditions associated with  $r(\omega)$  and  $t(\omega)$ . Determination of the optical field.

Assuming vaidity of quasimode scheme one determines the optical field (i.e. a(t)) inside the cavity by a Fano diagonalization precedure.

#### **DETERMINATION OF COUPLING CONSTANT** $V(\omega)$

The comparison with the exact solutions of the Maxwell equations in the limit of high reflectivity of the mirrors (DBR) leads to:

$$|V(\omega)|^2 = \frac{v}{4\pi L'} |t(\omega)|^2$$
$$-\frac{F(\omega) L'}{v} = \arg(r(\omega)) \equiv \varphi(\omega)$$

Moreover the Kramers - Kronig relations between  $\varphi(\omega)$  and  $t(\omega)$  in the limit of high reflectivity are recovered in this quasimode scheme

$$\varphi(\omega) = -\frac{1}{2\pi} \wp \int_{-\infty}^{+\infty} d\omega' \frac{|t(\omega')|^2}{\omega - \omega'}$$

$$\left[\varphi(\omega) = \frac{2\omega}{\pi} \wp \int_{0}^{+\infty} d\omega' \frac{\ln |r(\omega')|}{\omega^{2} - {\omega'}^{2}}\right]$$

## POLARITONS IN AN OPEN CAVITY. QUASIMODE SCHEME

$$H_{\rm int} = \int d\omega \left\{ V^{*}(\omega) a^{+} [b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega)] + h.c. \right\}$$

For normal incidence (q = 0),  $k_z = 2\pi/L'$ , and considering one cavity mode only (two polariton modes) the photon operator in terms of polaritons reads

$$a^{+} = W_{k_{t}}(\omega_{1}) B^{1+} + W_{k_{t}}(\omega_{2}) B^{2+}$$
$$-Y_{k_{t}}^{*}(\omega_{1}) B^{1} - Y_{k_{t}}^{*}(\omega_{2}) B^{2}$$

## Interaction between QW polaritons and the external radiation continuum

$$H_{\text{int}} = \int d\omega \sum_{i=1}^{2} \left\{ \tilde{V}_{i}^{*}(\omega) B^{i+} \left[ b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega) \right] + h.c. \right\}$$

$$+ \int d\omega \sum_{i=1}^{2} \left\{ \tilde{U}_{i}^{*}(\omega) B^{i} \left[ b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega) \right] + h.c. \right\}$$

$$\tilde{V}_{i}(\omega) = W_{k_{i}}^{*}(\omega_{i}) V(\omega) \quad \& \quad \tilde{U}_{i}(\omega) = -Y_{k_{i}}(\omega_{i}) V(\omega)$$

Close to resonance one has  $\left|\tilde{U}_{i}\left(\omega\right)\right|<<\left|\tilde{V}_{i}\left(\omega\right)\right|$ , the antiresonant terms can be realisted.

#### TIME EVOLUTION OF EXCITON STATES

$$\begin{split} H_{\text{int}} &= \int d\omega \sum_{i=1}^{2} \left\{ \tilde{V}_{i}^{*}(\omega) B^{i+} \left[ b_{\hat{\mathbf{k}}}(\omega) + b_{-\hat{\mathbf{k}}}(\omega) \right] + h.c. \right\} \\ A^{2} &\xrightarrow{t} A^{2}(t) = F_{1}(t) B^{1} + F_{2}(t) B^{2} \\ &+ \int d\omega \ f_{\hat{\mathbf{k}}}(\omega, t) \ b_{\hat{\mathbf{k}}}(\omega) + \int d\omega \ f_{-\hat{\mathbf{k}}}(\omega, t) \ b_{-\hat{\mathbf{k}}}(\omega) \end{split}$$

Initial condition: free exciton & no radiation.

The emission spectrum is related to  $F_1(t)$  and  $F_2(t)$ , which represent the probability amplitudes of finding a polariton in the evolved exciton state.

## In the Weisskopf - Wigner approximation the Fourier transforms take the form:

$$\left| \tilde{F}_1(\omega) \right|^2 = \frac{\Gamma_1/2}{\Gamma_1^2/4 + \left[\omega - (\omega_1 - \Delta\omega_1)\right]^2}$$
$$\left| \tilde{F}_2(\omega) \right|^2 = \frac{\Gamma_2/2}{\Gamma_2^2/4 + \left[\omega - (\omega_2 - \Delta\omega_2)\right]^2}$$

Linewidths  $\Gamma_1, \Gamma_2$  and frequency shifts  $\Delta\omega_1, \Delta\omega_2$  depend on the detuning (i.e. on L')

Complementarity of the semiclasical and quantum mechanical approach:

Sem	icla	ISS	ical
App	roa	ıch	

Exact solution of Maxwell equations with a linear non-local exciton response function.

Quantities like reflectivity, transmission, absorption can be directly obtained.

The nonradiative exciton line width is easily taken into account.

#### Quantum Approach

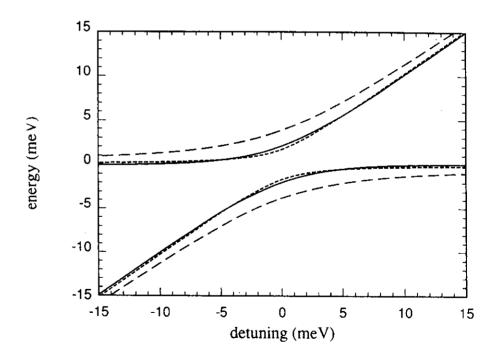
Solution of Heisenberg equations for polariton operators.
Some approximations are introduced.

Direct calculation of the luminescence spectrum.

Knowledge of the quantum states of the system at any given time.
Possibility to calculate the statistical properties of the radiation.

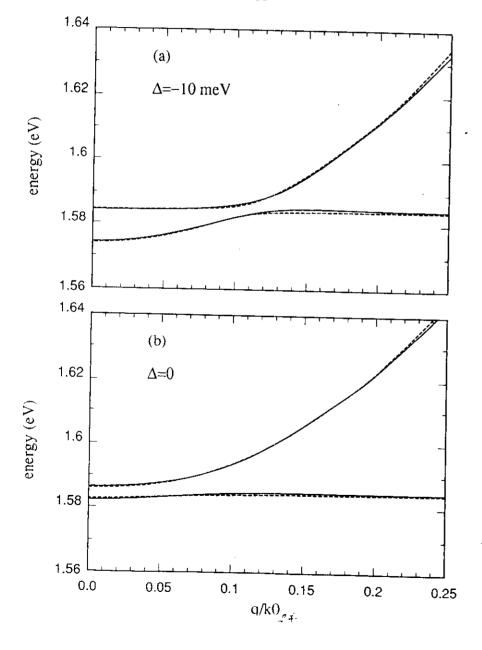
Comparison between the semiclassical and full quantum mechanical approaches for the polariton dispersion in a microcavity with losses as a function of the detuning.

Dashed line: closed cavity (classical and q. m) Dotted line: semiclassical approach Full line: quantum mechanical approach.



Comparison between the semiclassical and full quantum mechanical approaches for the polariton dispersion in a microcavity with losses as a function of the wave vector and for different detuning.

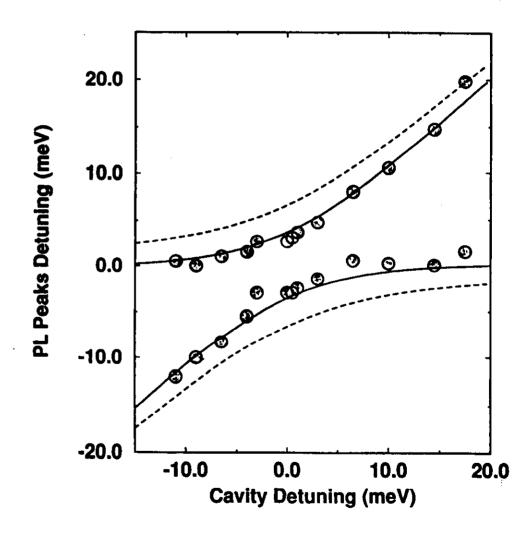
Dotted line: semiclassical approach Full line: quantum mechanical approach.



# EMISSION PEAK POSITION AS A FUNCTION OF THE DETUNING FOR A CAVITY WITH 5 QW

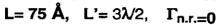
solid line: cavity with losses dotted line: closed cavity

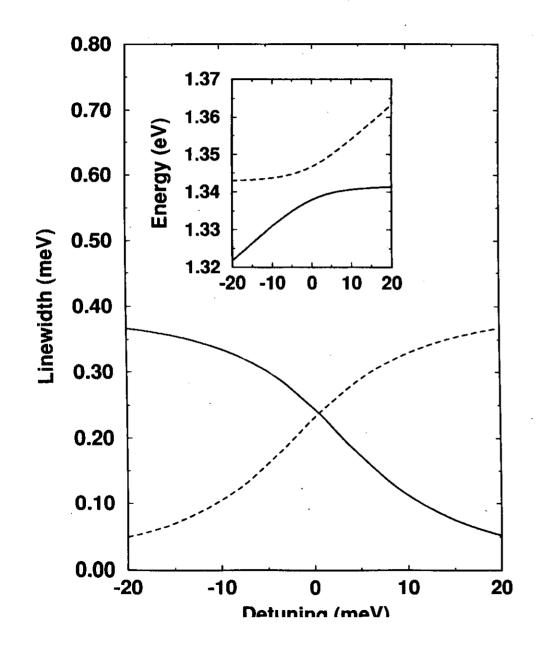
experiment: C. Weisbuch et al. Phys.Rev.Lett. 69,3314 (1992)

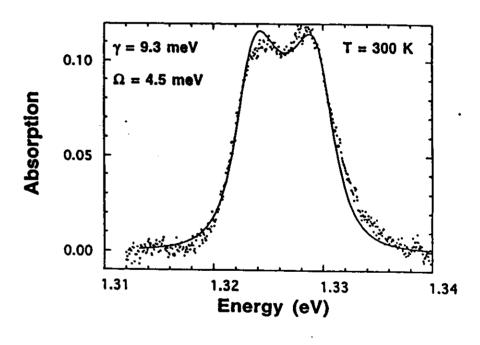


#### POSITION OF THE EMISSION PEAKS AND RADIATIVE LINEWIDTH AS A FUNCTION OF THE DETUNING

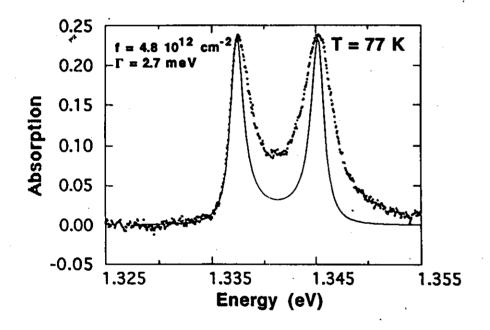
6 qw inside the cavity, exciton energy 1.3421 eV,







15



#### **EMISSION SPECTRUM**

Initial state: polaritons  $B^{1+}|0\rangle$  &  $B^{2+}|0\rangle$ 

Exciton component of a polariton:  $X_1$  &  $X_2$ 

$$B_{\mathbf{q}}^{l} = \sum_{i=1}^{l} W_{i}(k_{i}, \mathbf{q}) A_{\mathbf{q}, i_{i}, k}^{l} + X_{l}(\mathbf{q}) A_{\mathbf{q}}^{2}$$

$$+\sum_{i=1}^{n}Y_{i}(k_{i},\mathbf{q})\ A_{-q,k_{i},k}^{i^{*}}+Z_{i}(\mathbf{q})\ A_{-q}^{i^{*}}$$

Boltzmann factors:

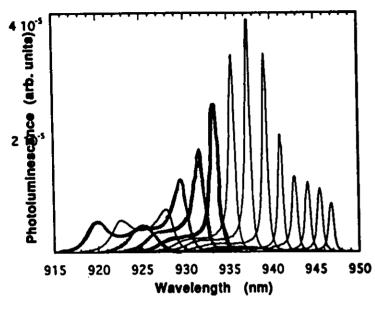
$$\exp[-\beta\hbar\omega_1]/N$$
 &  $\exp[-\beta\hbar\omega_2]/N$ 

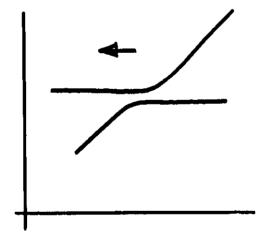
$$N = \exp[-\beta\hbar\omega_1] + \exp[-\beta\hbar\omega_2]$$

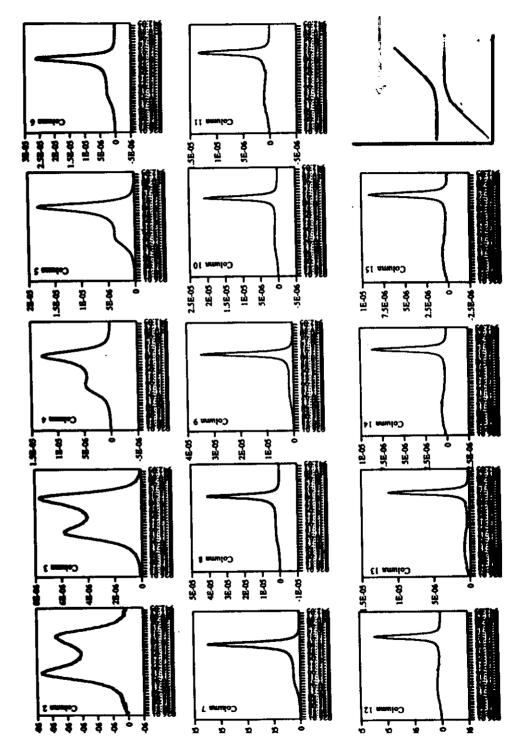
Final states: radiation states  $b_k^*(\omega)|0\rangle$ 

#### **Emission spectrum**

$$E(\omega) = \left| \langle 0 | \mathbf{b}_{\hat{\mathbf{k}}}(\omega) B^{1+} | 0 \rangle \right|^2 \left| X_1 \right|^2 \exp \left[ -\beta \hbar \omega_1 \right] / N$$
$$+ \left| \langle 0 | \mathbf{b}_{\hat{\mathbf{k}}}(\omega) B^{2+} | 0 \rangle \right|^2 \left| X_2 \right|^2 \exp \left[ -\beta \hbar \omega_2 \right] / N$$





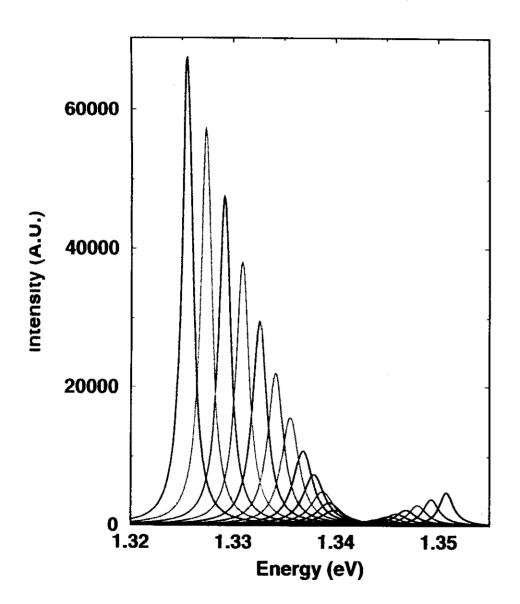


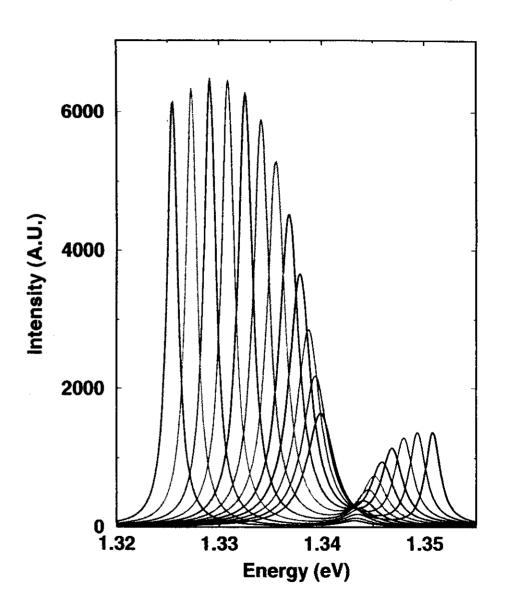
#### **LUMINESCENCE**

6 qw inside the cavity, exciton energy 1.3421 eV, L= 75 Å, L'= 3 $\lambda$ /2,  $\Gamma_{n.r.}$ = 4 meV, T= 110 K

#### LUMINESCENCE

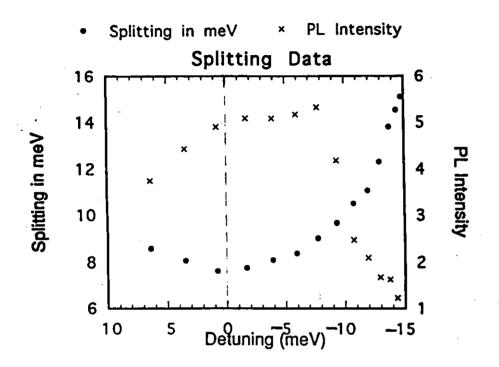
6 qw inside the cavity, exciton energy 1.3421 eV, L= 75 Å, L'=  $3\lambda 2$ ,  $\Gamma_{\text{n.r.}}$ = 4 meV, T= 110 K

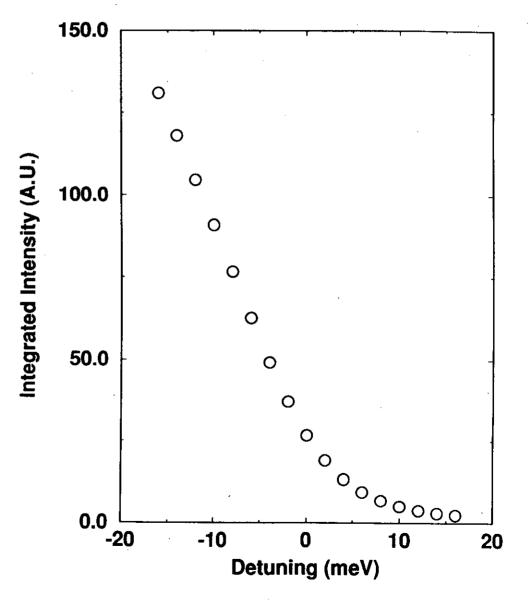




#### **LUMINESCENCE: INTEGRATED INTENSITY**

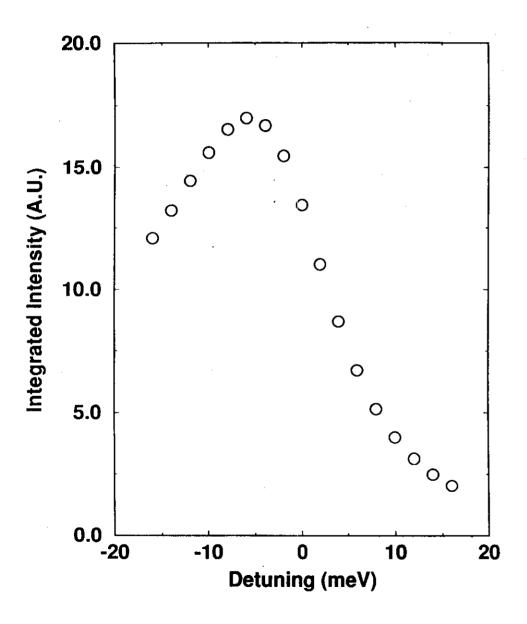
6 qw inside the cavity, exciton energy 1.3421 eV, L= 75 Å, L'= 3 $\lambda$ /2,  $\Gamma_{n.r.}$ = 4 meV, T= 110 K





#### **LUMINESCENCE: INTEGRATED INTENSITY**

6 qw inside the cavity, exciton energy 1.3421 eV, L= 75 Å, L'=  $3\lambda/2$ ,  $\Gamma_{n.r.}$ = 4 meV, T= 110 K



#### REMARKS

- \* The restriction to one cavity mode only is an excellent approximation.
- \* The quasimode approximation is only justified in the limit of high reflectivity of the mirrors.
- \* The model has to be extended to include non normal incidence and different light polarizations.
- \* The model does not contain the nonradiative lifetime of the exciton.
- \* The model allows to calculate statistical properties of the radiation field.
- \* The model has to be extended in order to describe PL results.

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