



# Four-Wave Mixing and Many-Particle Effects in Semiconductors

**Rolf Binder**

College of Optical Sciences and Department of Physics  
The University of Arizona



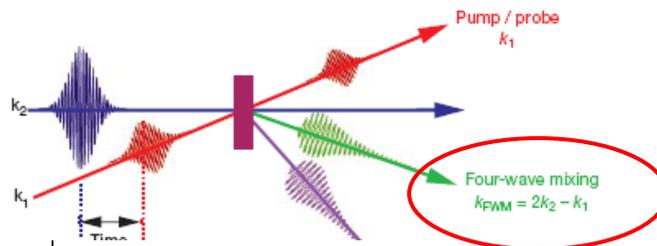
# Many-body and correlation effects in semiconductors

D. S. Chemla\* & Jagdeep Shah†

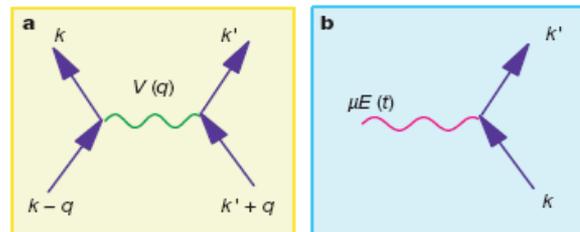
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† Bell Laboratories, Lucent Technologies, Holmdel, New Jersey 07733, USA

## Box 1 Principle of time-resolved nonlinear optical spectroscopy experiments



## Box 2 Many-body effects in light-semiconductor interaction



Box 2 Figure Feynman diagrams for (a) Coulomb interaction between carriers, and (b) coupling with electromagnetic field.

Box 1 Figure Schematic of a generic time-reso



phys. stat. sol. (b) **221**, 195 (2000)

Subject classification: 71.35.Gg; 78.47.+p; S7.12

## Formation and Decay of Coherent Four-Particle Correlations in Semiconductors: A Green's Function Theory

W. SCHÄFER (a), R. LÖVENICH (a), N. FROMER (b), and D. S. CHEMLA (b)

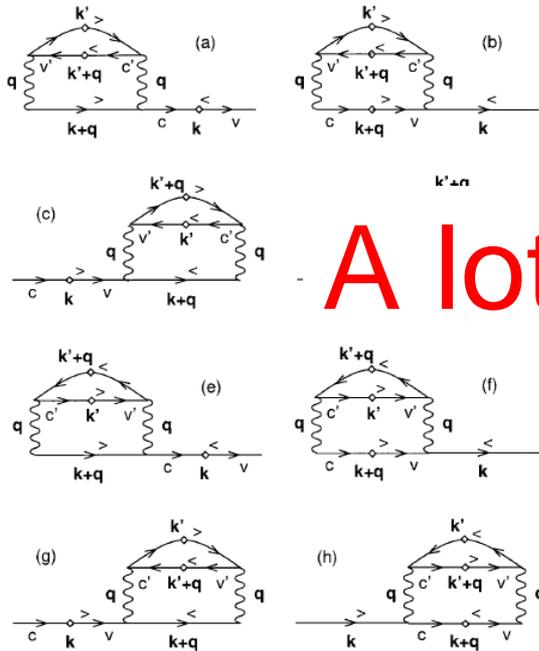


Fig. 1. Diagrammatic representation of the RPA polarization scattering diagrams in second-order Born approximation

**A lot of diagrams...**

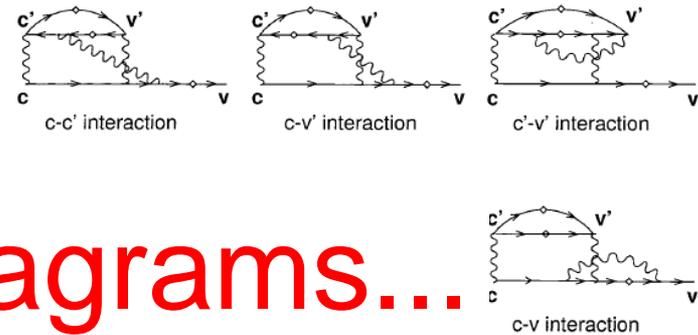


Fig. 2. Third-order contributions resulting from the RPA polarization scattering diagram a) of Fig. 1

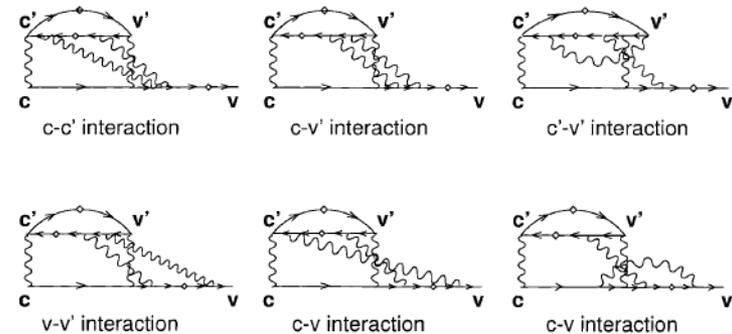


Fig. 3. Fourth-order contributions resulting from the second diagram of Fig. 2



This tutorial is about

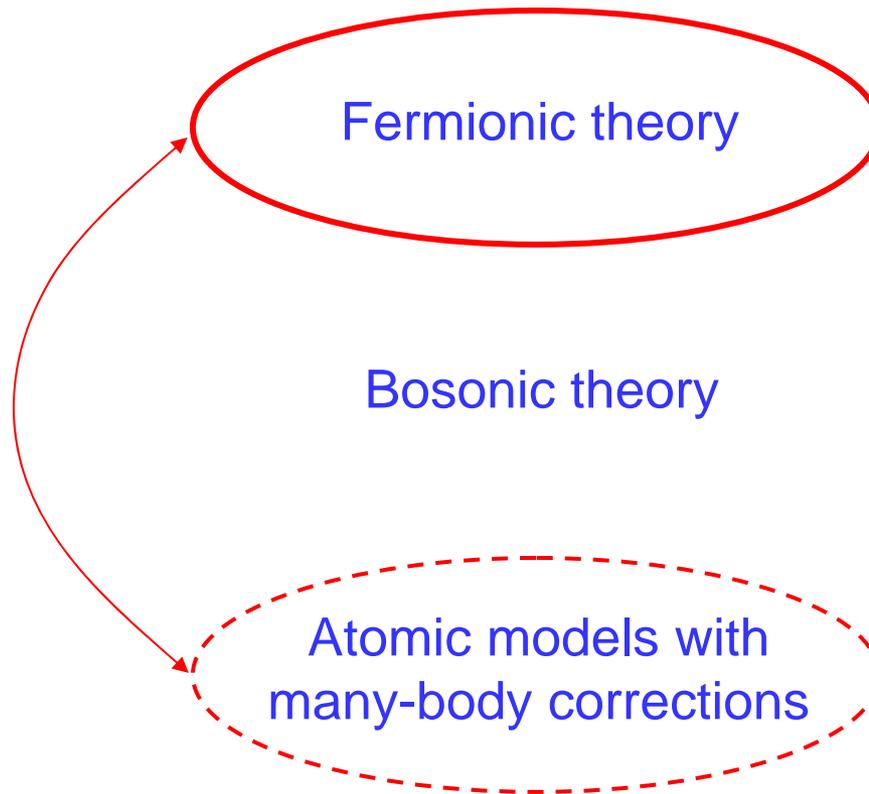
using Feynman diagrams and four-wave mixing to study many-particle correlations in semiconductors

It is **not** about

photon echo, holography, phase conjugation, third-harmonic generation, ...



## Three theoretical approaches





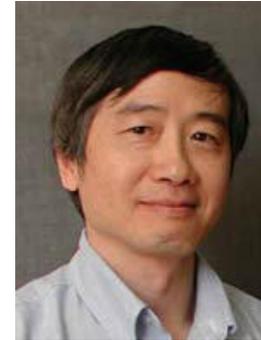
*First:*

Acknowledgements



## Special thanks to

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College of Optical Sciences  
University of Arizona



### ...and to (past and present) graduate students and postdocs

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Dan Nguyen (now NP Photonics)

Stefan Schumacher (now Heriot-Watt Univ.)

Greg Rupper

Baijie Gu



ERATO, AFOSR, DARPA, JSOP



Outline:

- Introduction
- Many-particle theory and Green's functions
- Third-order optical response
- Correlations beyond third order
- Few-level systems
- FWM instabilities (time permitting)



*Next:*

Weekend experiences with  
perturbative and non-perturbative  
two-particle correlations

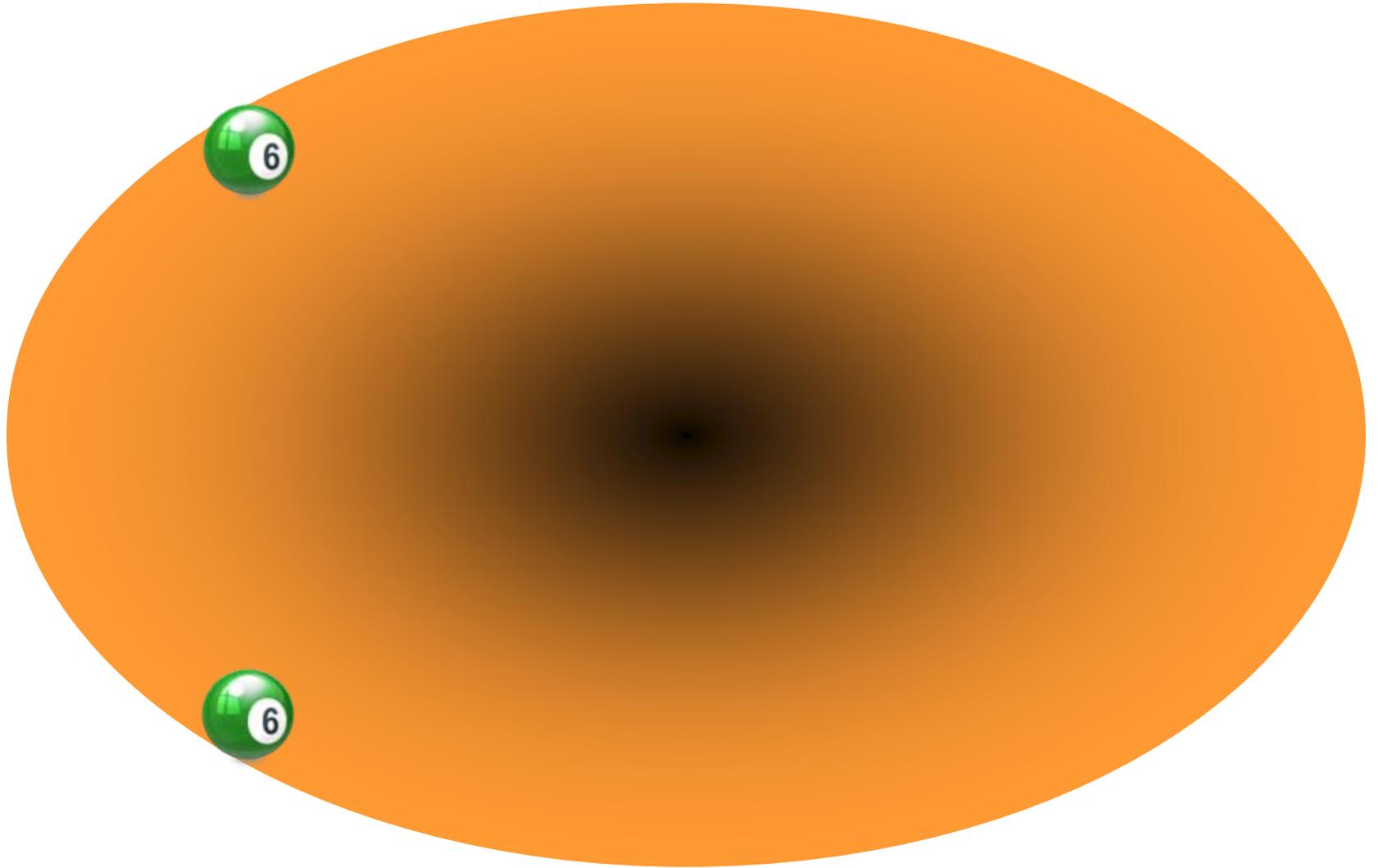


## Two-particle interaction





## Two-particle correlations: bound states





## Two-particle correlation: continuum states





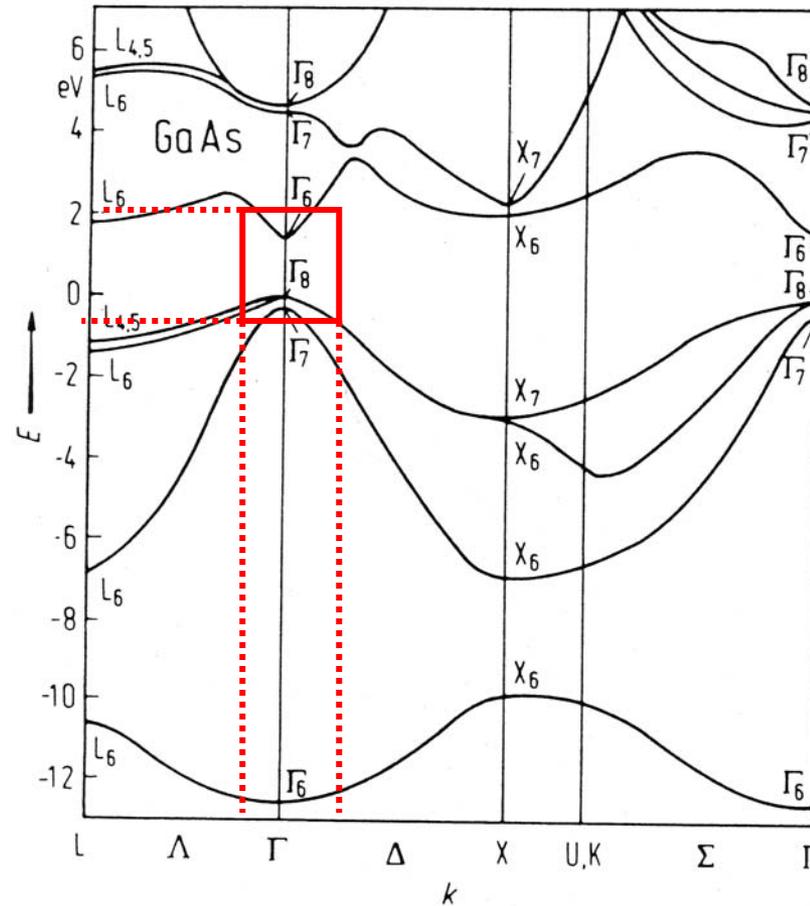
*Next:*

Introduction to semiconductors



## GaAs bandstructure

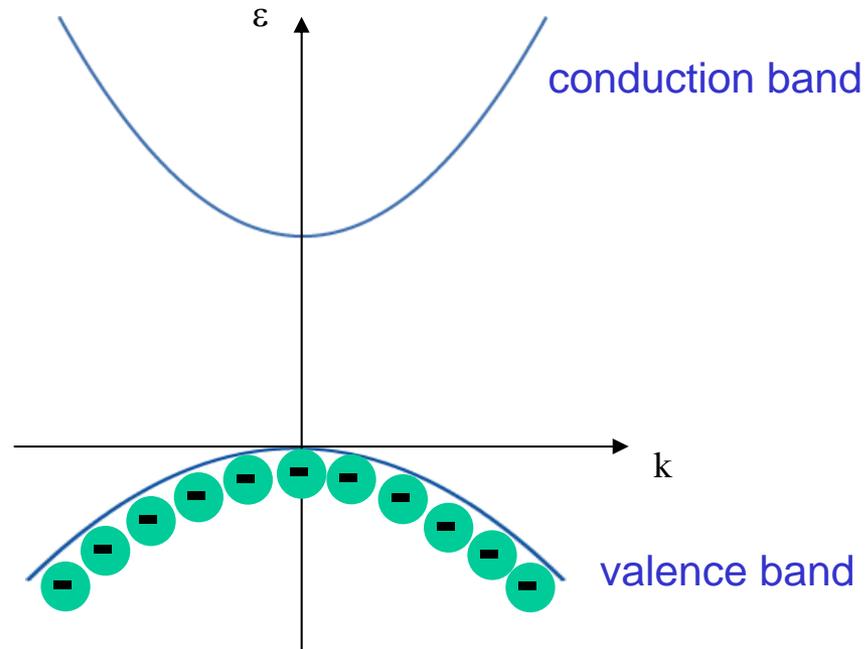
Optical excitation  
typically close to  
 $\Gamma$  point ( $k=0$ )





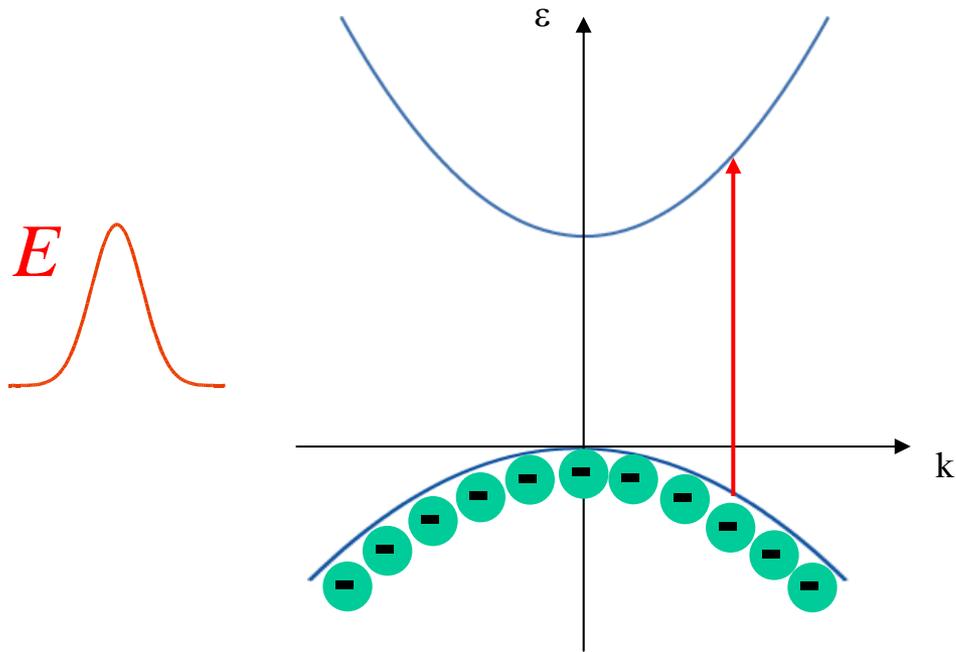
## Parabolic Bandstructure near $k=0$

Ground state:  
full valence band



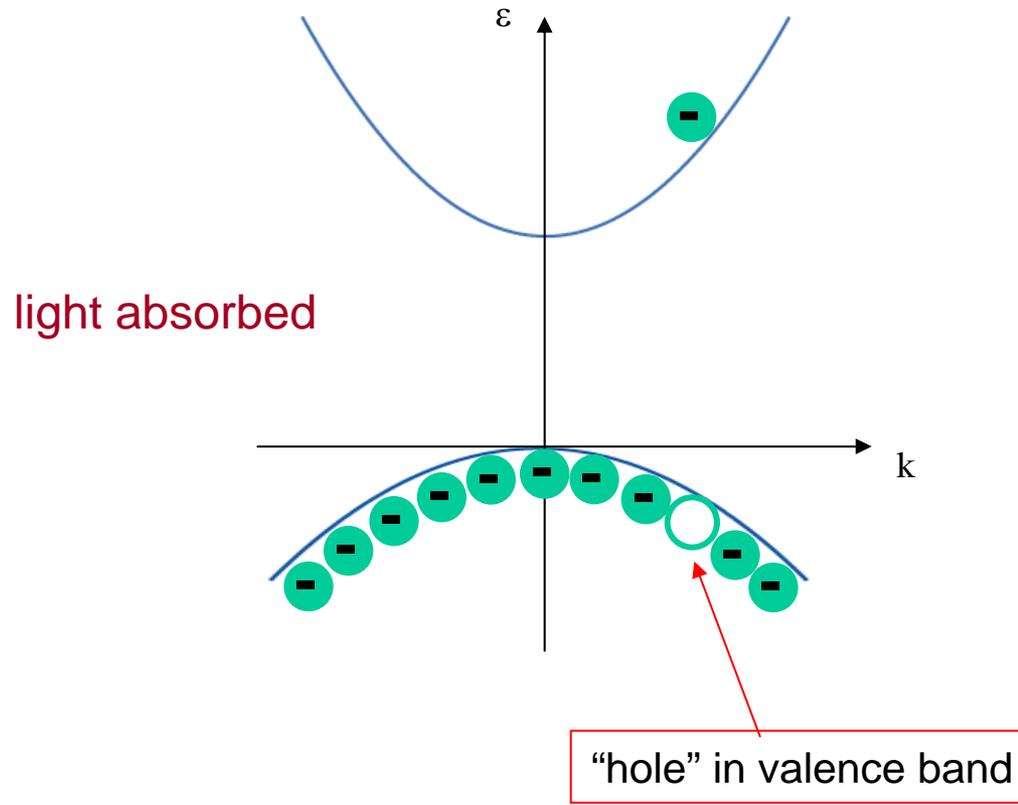


## Optical excitation



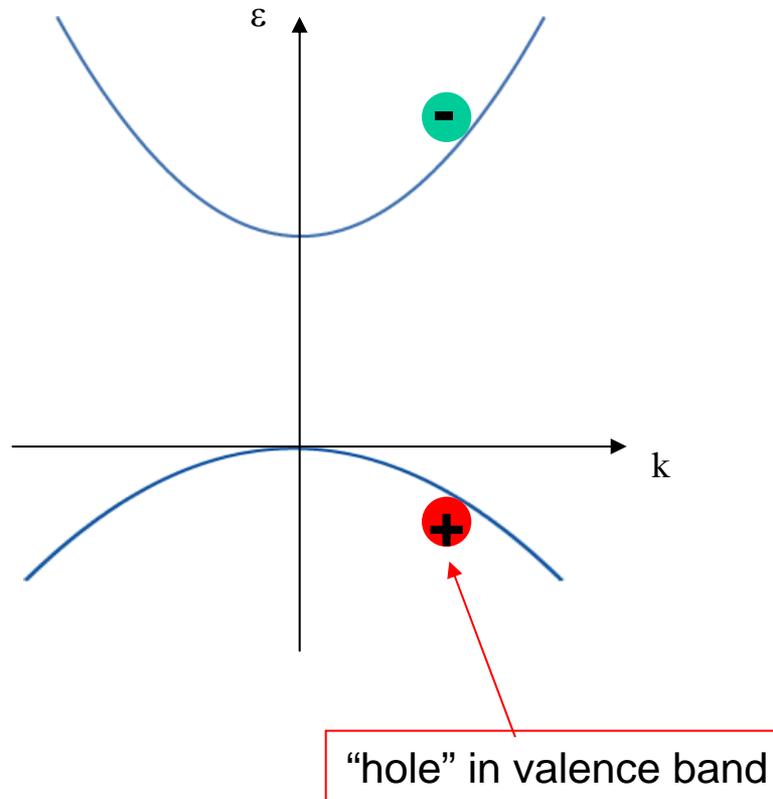


## Optical excitation



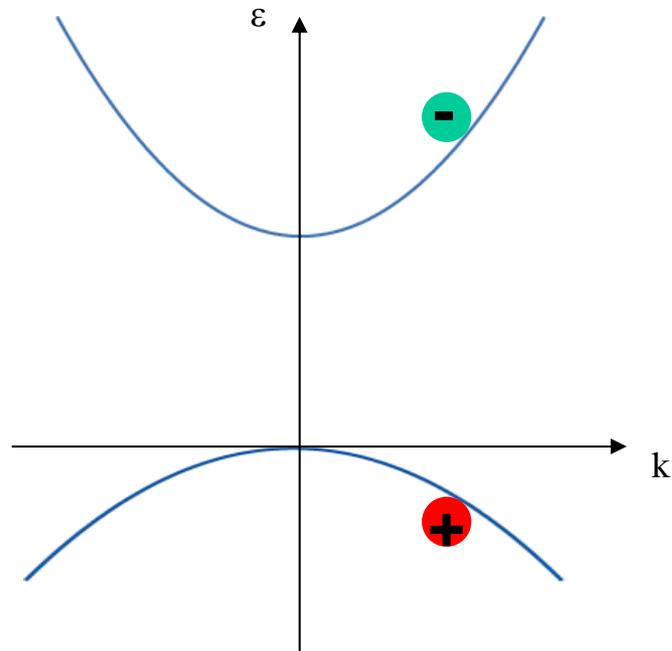


Hole = positively charged “particle”



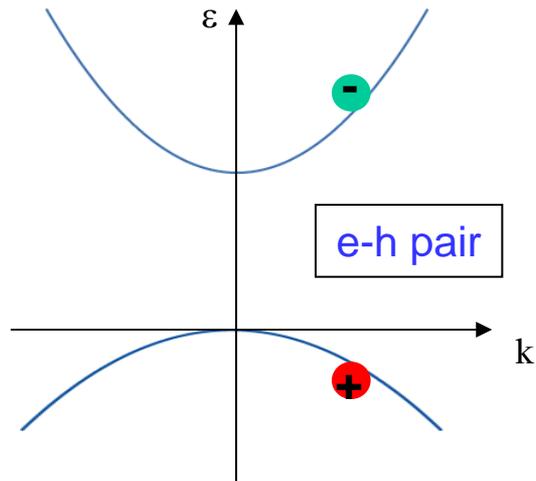


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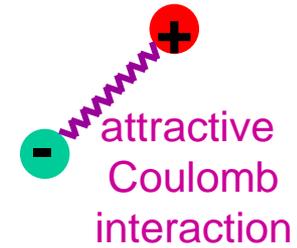




## Excitons



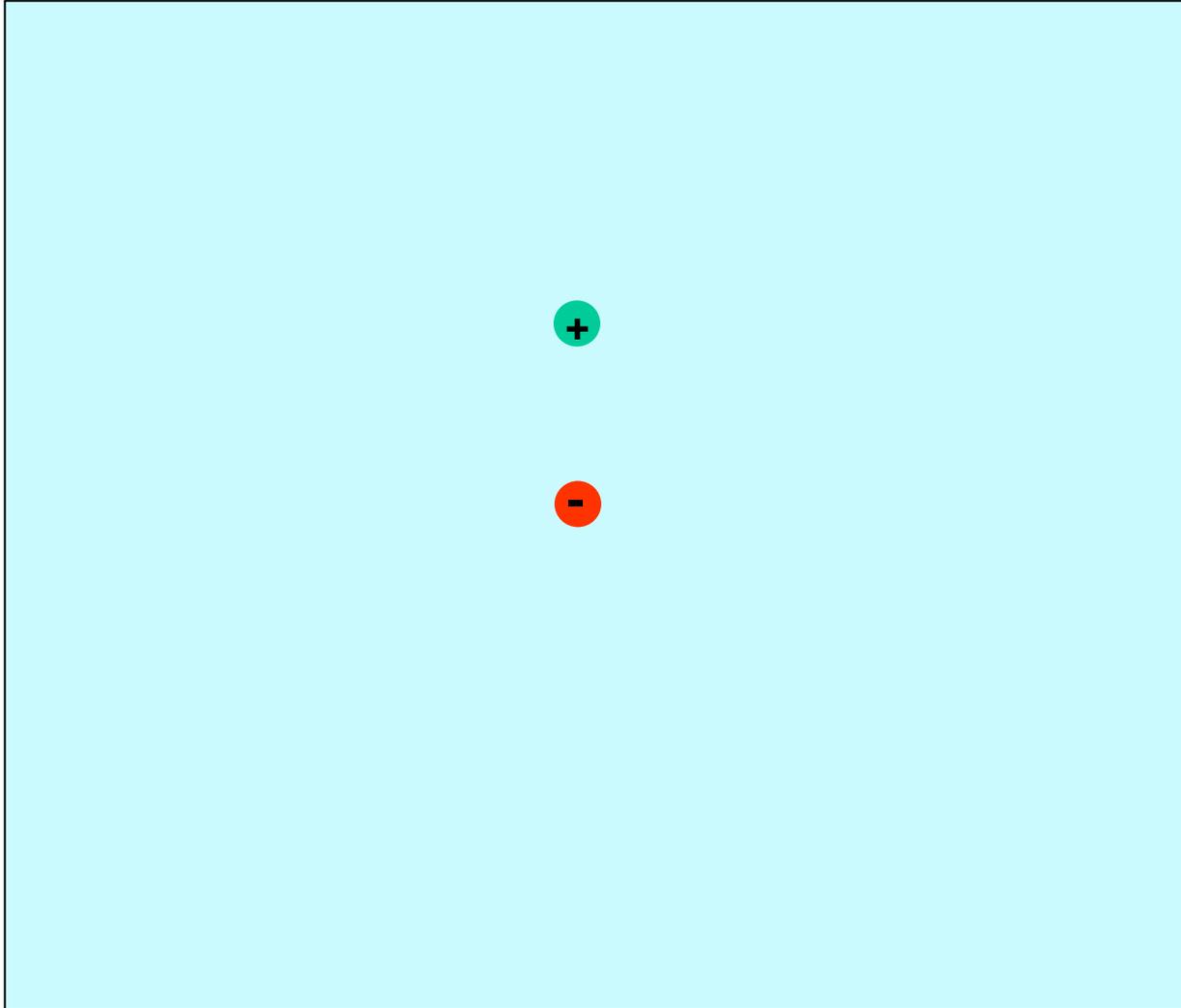
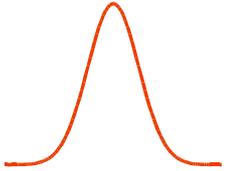
real space:



- ◆ Bound states (Rydberg law)  $\epsilon_n = E_g - \frac{E_b}{n^2}$
- ◆ Continuum states



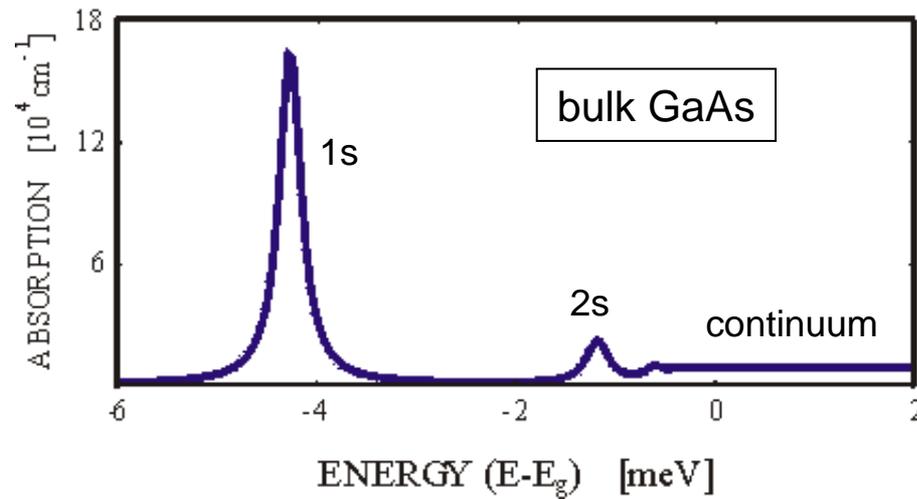
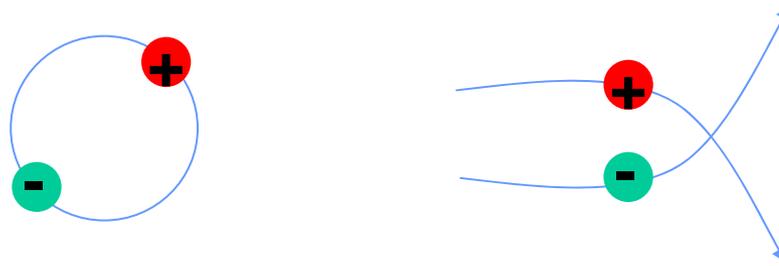
# Excitons





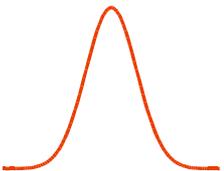
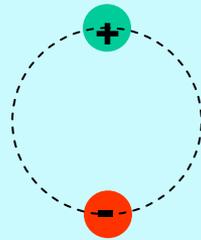
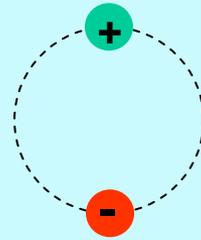
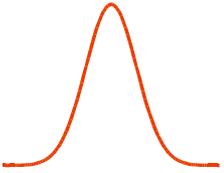
# Excitons

real space:





## Nonlinear excitation: two-exciton states



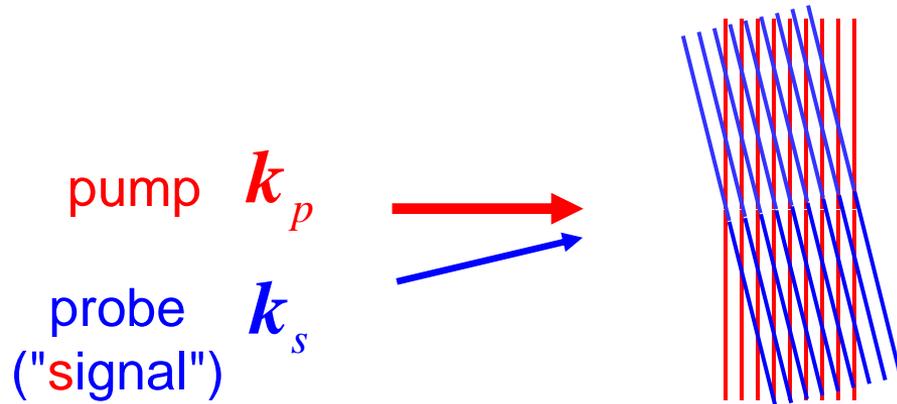


*Next:*

The concept of four-wave mixing (FWM)



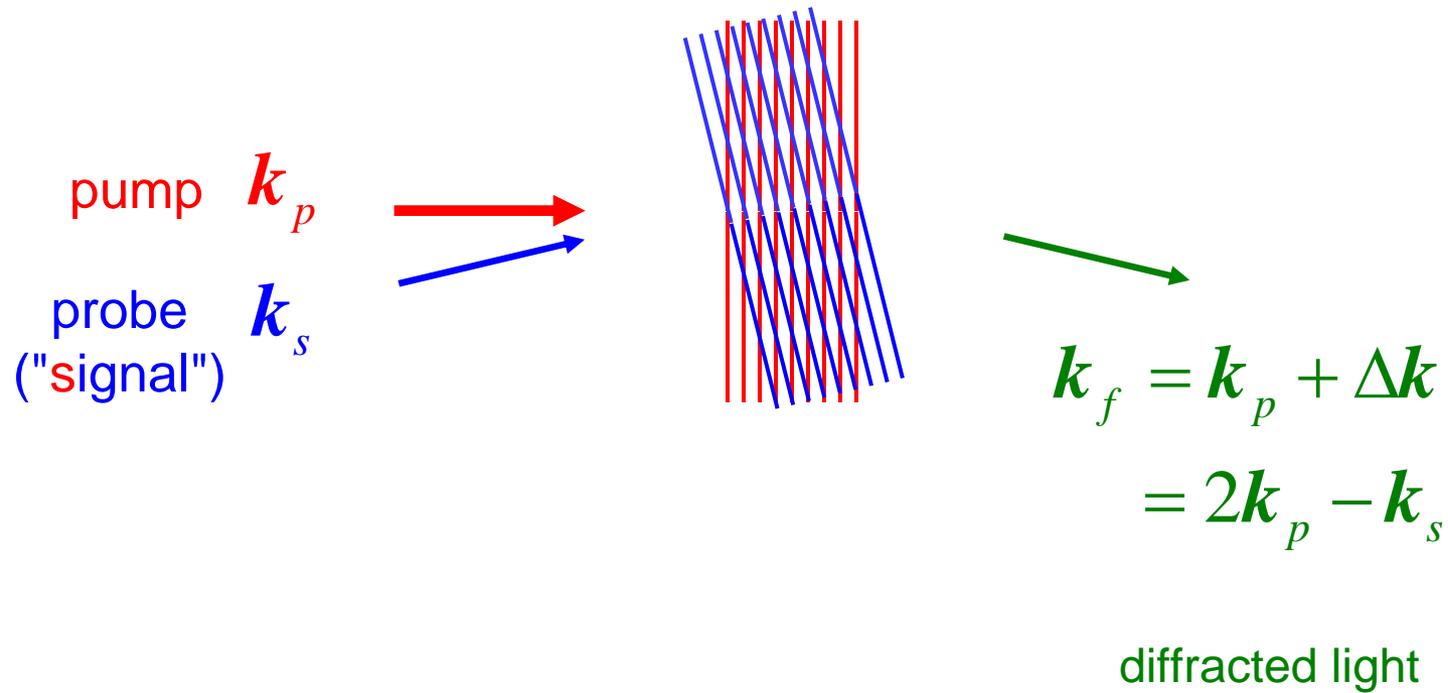
## Wave mixing and self diffraction



Grating with spatial wave vector  $\Delta \mathbf{k}_f = \mathbf{k}_p - \mathbf{k}_s$



## Forward four-wave mixing





$$\left\{ \frac{\epsilon_b}{c^2} \frac{\partial^2}{\partial t^2} - \nabla^2 \right\} \mathbf{E} = -\frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}$$

Third-order response:  
(schematically; integrals over space  
and time suppressed)

$$P = \chi^{(3)} E^* E E$$

spatial  
dependence:

$$e^{ik_f r}$$

$$e^{-ik_s r}$$

$$e^{ik_p r}$$

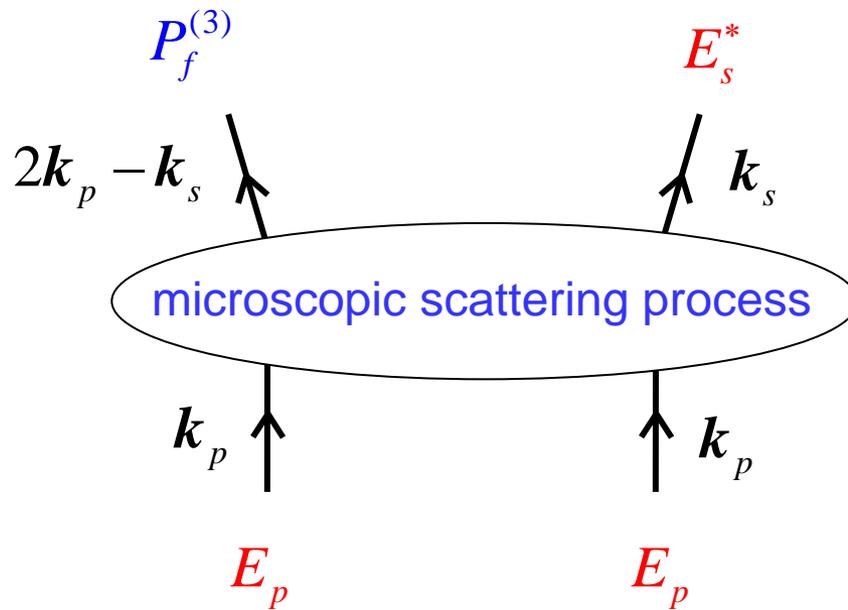
$$e^{ik_p r}$$

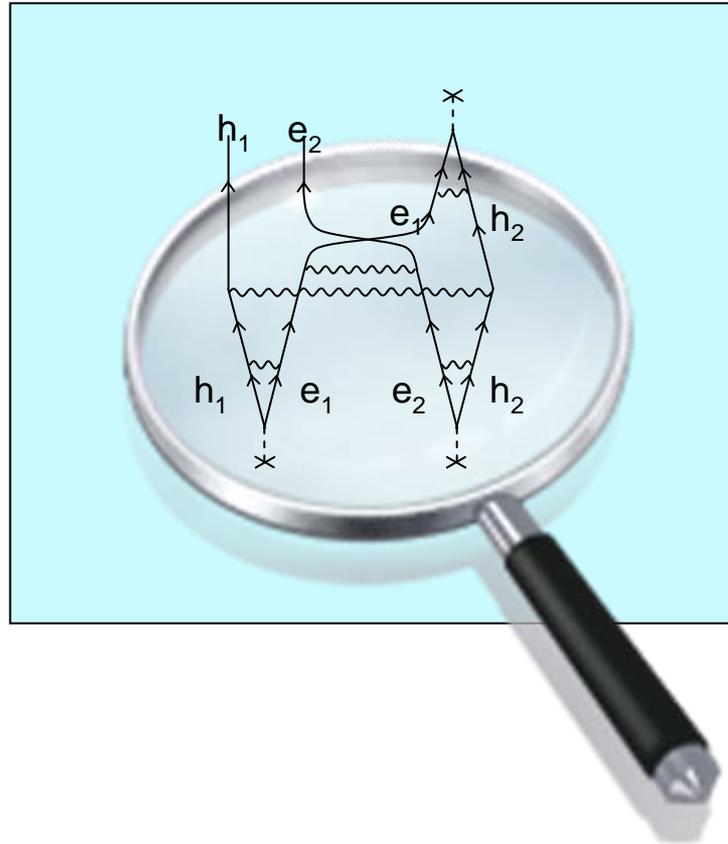
information about excitonic correlations

Strategy in this talk: present P as Feynman diagrams



## Four-wave mixing





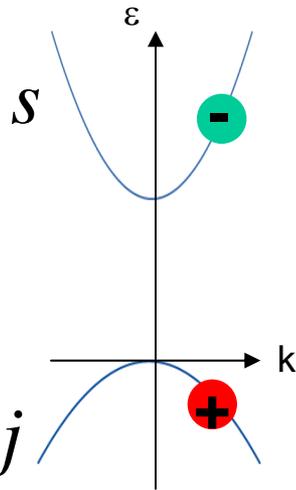


*Next:*

Introduction to many-particle theory



electrons



holes

$$a_{sk}^\dagger$$

$$a_{sk}$$

$$a_{jk}^\dagger$$

$$a_{jk}$$

creation  
operators

annihilation  
operators



## Fermionic semiconductor Hamiltonian

$$H = H_{band} + H_{Coulomb} + H_{light-coupling}$$

$$H_{band} = \sum_{sk} \varepsilon_{sk}^e \underbrace{a_{sk}^\dagger a_{sk}}_e + \sum_{jk} \varepsilon_{jk}^h \underbrace{a_{jk}^\dagger a_{jk}}_h$$

e and h occupation number operators

$$H_{Coulomb} = \frac{1}{2} \sum_{\text{all indices}} V_q^c \left[ \underbrace{a_{s,k+q}^\dagger a_{s',k-q}^\dagger a_{s',k'} a_{s,k}}_{e-e} + \underbrace{a_{j,k+q}^\dagger a_{j',k-q}^\dagger a_{j',k'} a_{j,k}}_{h-h} + 2 \underbrace{a_{s,k+q}^\dagger a_{j,k-q}^\dagger a_{j,k'} a_{s,k}}_{e-h \text{ interaction}} \right]$$

$$H_{light-coupling} = \sum_{\text{all indices}} \left[ \underbrace{\vec{\mu}_{sj}^* \cdot \vec{E}^*(t) a_{s,k} a_{j,-k}}_{\text{electron-hole pair annihilation}} + \underbrace{\vec{\mu}_{sj} \cdot \vec{E}(t) a_{s,k}^\dagger a_{j,-k}^\dagger}_{\text{electron-hole pair creation}} \right]$$

electron-hole pair annihilation and creation operators

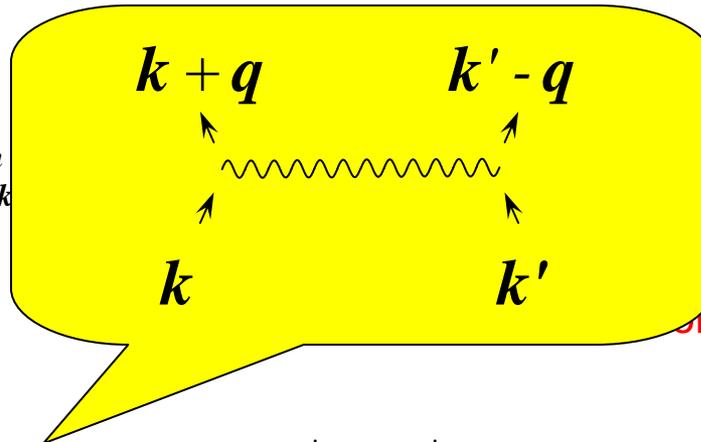


## Fermionic semiconductor Hamiltonian

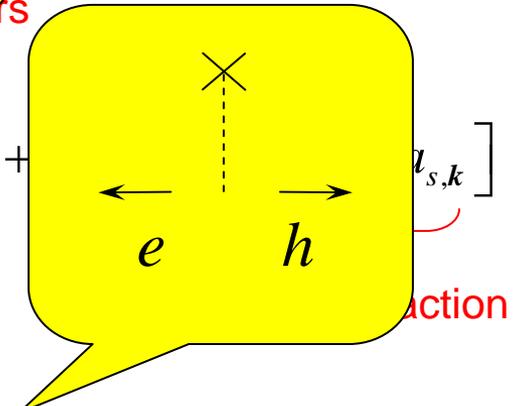
$$H = H_{band} + H_{Coulomb} + H_{light-coupling}$$

$$H_{band} = \sum_{sk} \varepsilon_{sk}^e \underbrace{a_{sk}^\dagger a_{sk}}_e + \sum_{jk} \varepsilon_{jk}^h \underbrace{a_{jk}^\dagger a_{jk}}_h$$

e and h states



$$H_{Coulomb} = \frac{1}{2} \sum_{\text{all indices}} V_q^c \left[ \underbrace{a_{s,k+q}^\dagger a_{s',k-q}^\dagger a_{s',k'} a_{s,k}}_{e-e} + \underbrace{a_{j,k+q}^\dagger a_{j',k-q}^\dagger a_{j',k'} a_{j,k}}_{h-h} + \underbrace{a_{s,k}^\dagger a_{s',k}^\dagger a_{s',k'} a_{s,k'}}_{e-h \text{ interaction}} \right]$$



$$H_{light-coupling} = \sum_{\text{all indices}} \left[ \underbrace{\vec{\mu}_{sj}^* \cdot \vec{E}^*(t) a_{s,k} a_{j,-k}}_{\text{annihilation}} + \underbrace{\vec{\mu}_{sj} \cdot \vec{E}(t) a_{s,k}^\dagger a_{j,-k}^\dagger}_{\text{creation}} \right]$$

electron-hole pair annihilation and creation operators



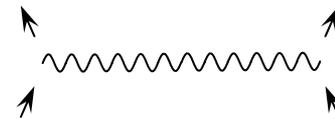
$$H = H_{band} + H_{Coulomb} + H_{light-coupling}$$

**Show only basic structure:**

$$H_{band} = \varepsilon a^\dagger a$$

$$H_{Coulomb} = V^c a^\dagger a^\dagger a a$$

$$H_{light-coupling} = E^* a a + E a^\dagger a^\dagger$$





*Next:*

About expectation values and Green's functions



## Expectation values

$$p_{eh}(t) = \langle a_h(t) a_e(t) \rangle$$

$$f_e(t) = \langle a_e^\dagger(t) a_e(t) \rangle$$

$$f_h(t) = \langle a_h^\dagger(t) a_h(t) \rangle$$

*interband polarization*

*occupation functions*

## Two-time functions

$$p_{eh}(t) = \langle a_h(t) a_e(t') \rangle \Big|_{t=t'}$$

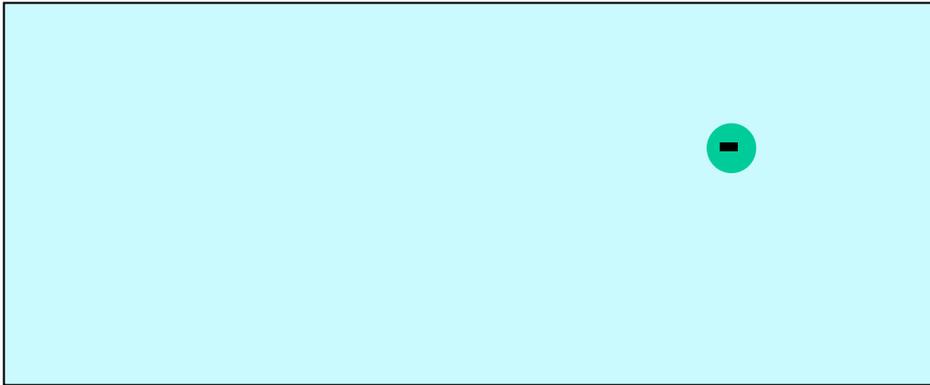
$$f_e(t) = \langle a_e^\dagger(t) a_e(t') \rangle \Big|_{t=t'}$$



## Particle propagators

$$\langle a(t) a^\dagger(t') \rangle$$

$t$  later than  $t'$



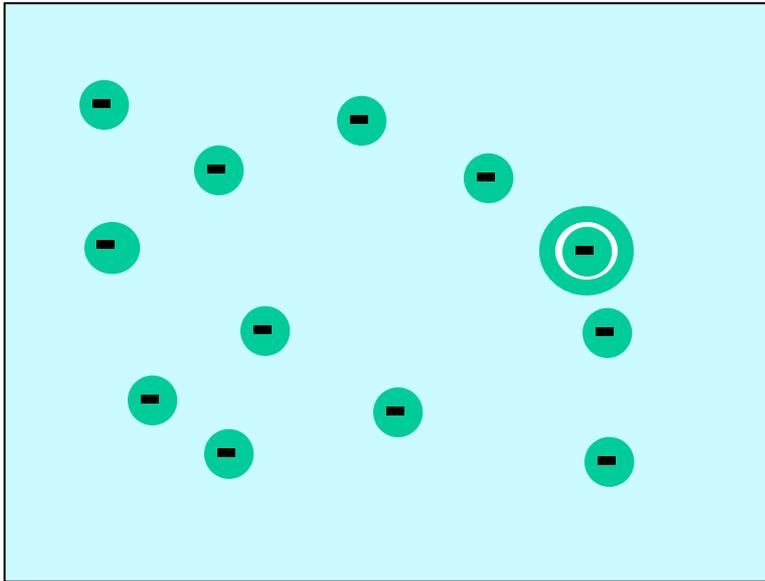
carries information about particle energy



## Hole propagators

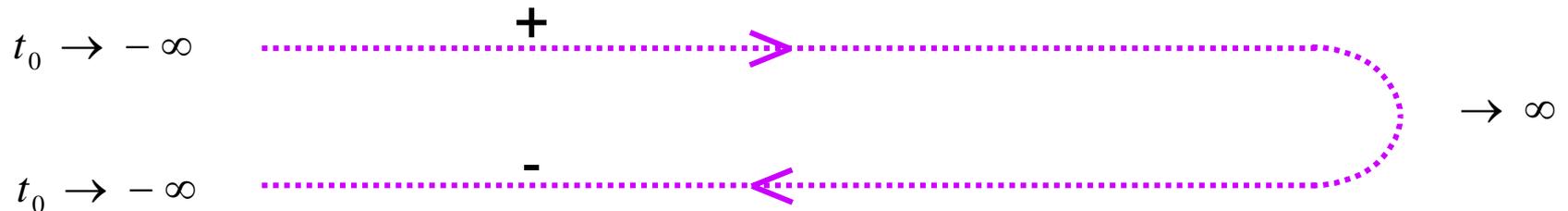
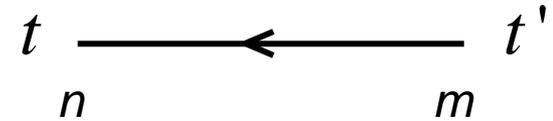
$$\langle a^\dagger(t') a(t) \rangle$$

$t'$  later than  $t$



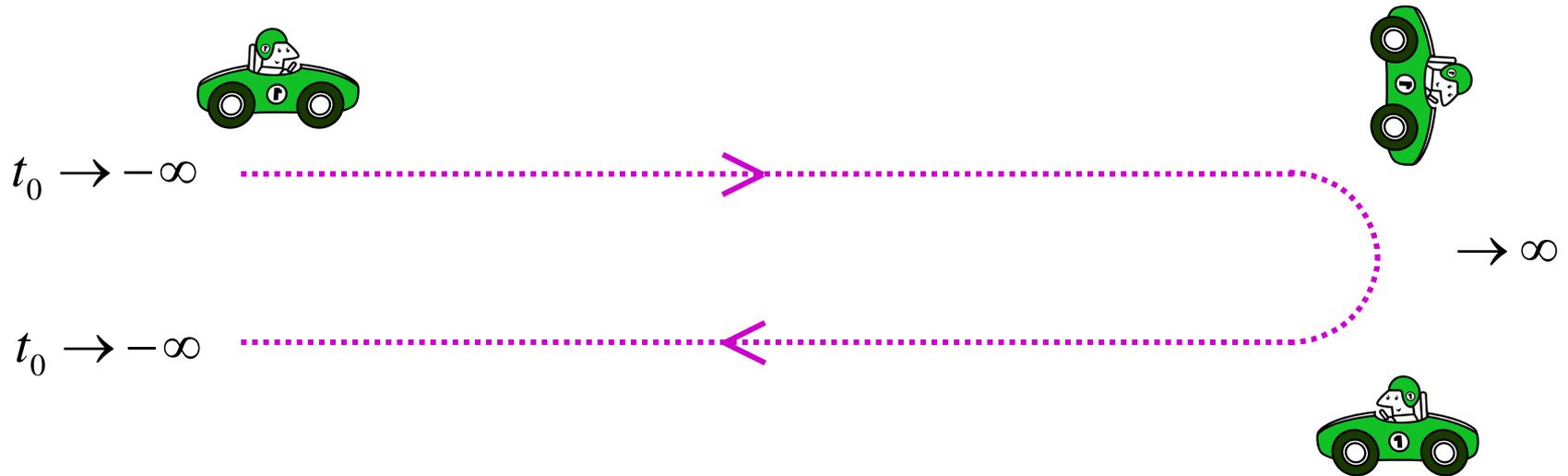


$$iG_{nm}(t, t') = \langle \hat{T}_c a_n(t) a_m^\dagger(t') \rangle$$



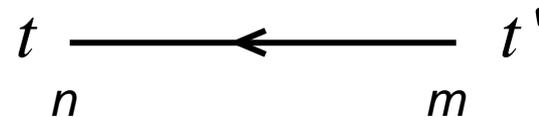
if  $t'$  **later** on contour than  $t$ : 
$$iG_{nm}(t, t') = -\langle a_m^\dagger(t') a_n(t) \rangle$$

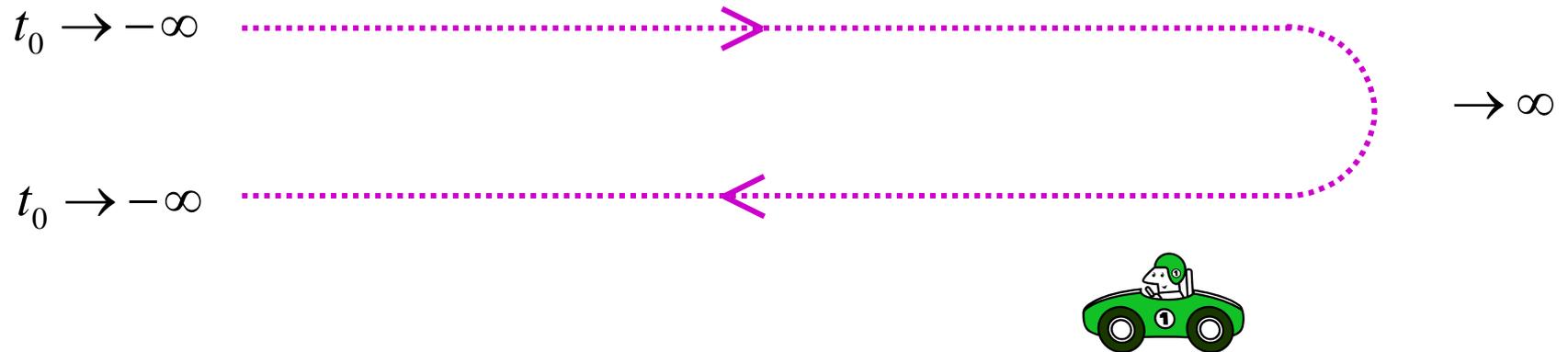
if  $t'$  **earlier** on contour than  $t$ : 
$$iG_{nm}(t, t') = +\langle a_n(t) a_m^\dagger(t') \rangle$$



All one-particle Green's functions have "time arrow"

$$iG_{nm}(t, t') = \langle \hat{T}_c a_n(t) a_m^\dagger(t') \rangle$$





Usual propagators (non density-type)

$t \longleftarrow t'$   
arrow points **forward** in time

$$iG_{nm}(t, t') = \langle a_n(t) a_m^\dagger(t') \rangle$$



"Density-type"

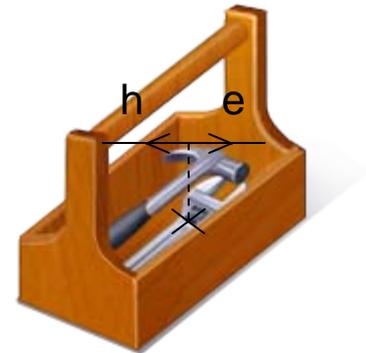
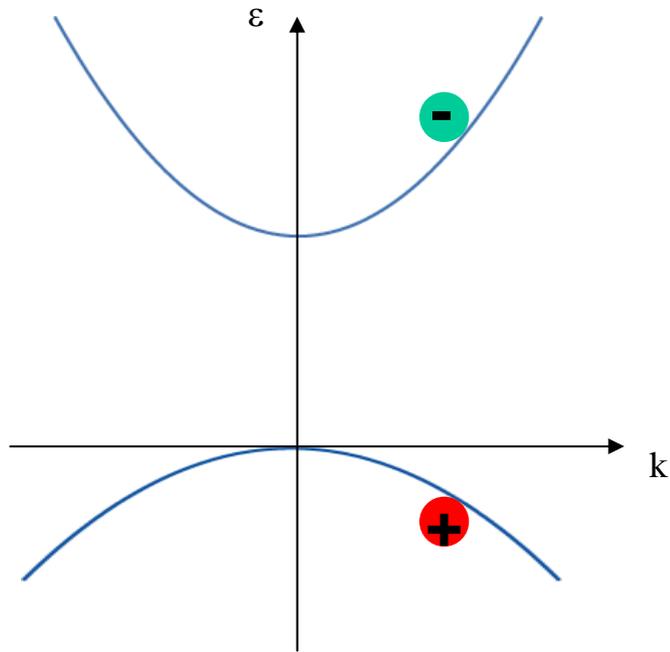
$t \longleftarrow t'$   
arrow points **backward** in time

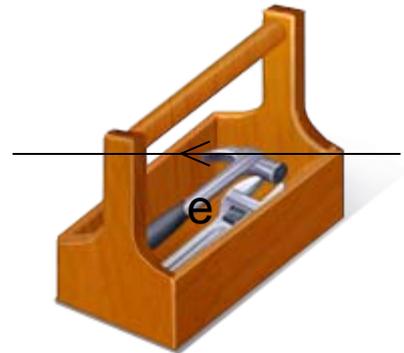
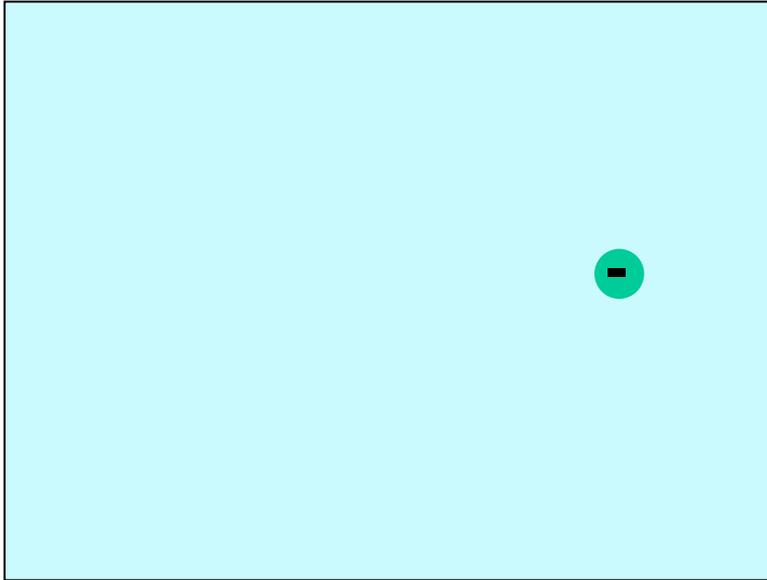
$$iG_{nm}(t, t') = \langle a_m^\dagger(t') a_n(t) \rangle$$

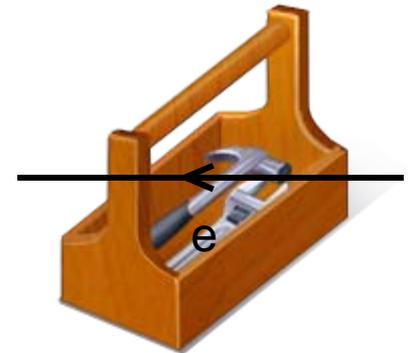
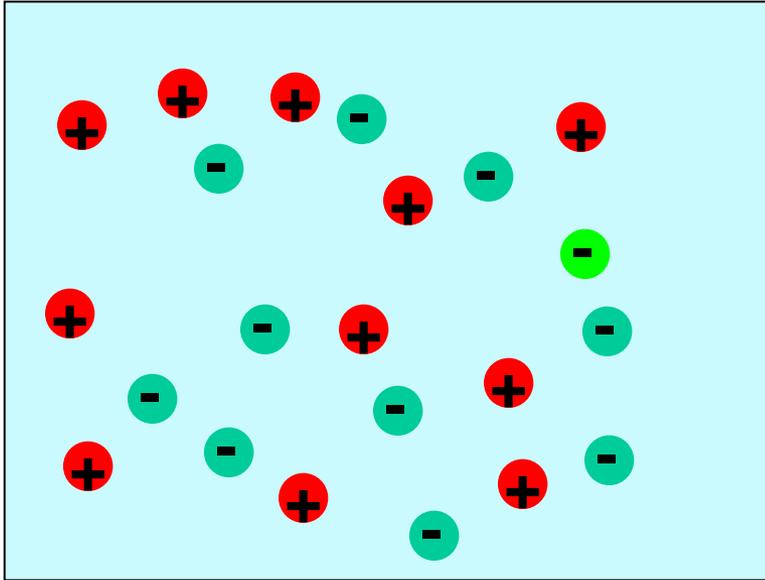


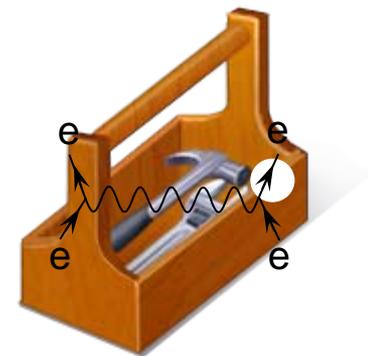
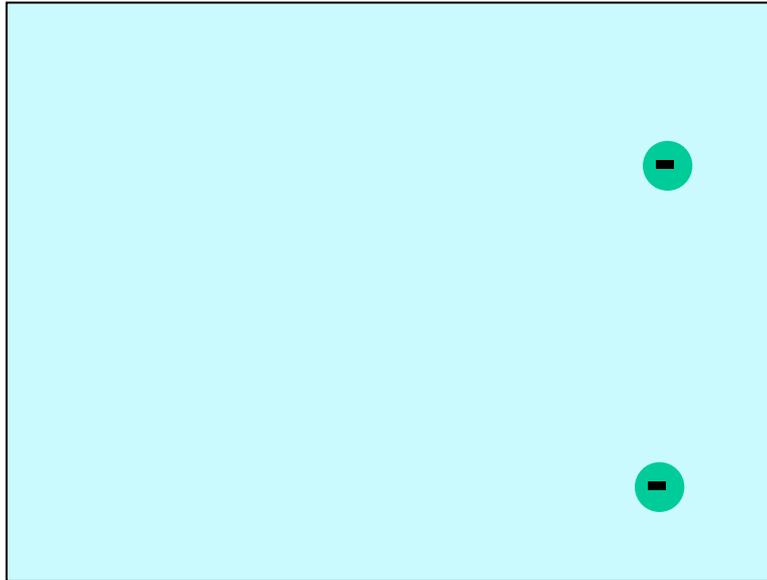
*Next:*

A diagram tool box











$\nu_1, \vec{k} + \vec{q}$        $\nu_2, \vec{k}' - \vec{q}$   
 $\left\{ \begin{matrix} \vec{t}_1, j, \vec{k} \\ \vec{t}_1, j, \vec{k} \end{matrix} \right\}$        $\left\{ \begin{matrix} \vec{t}_2, s, \vec{k}' \\ \vec{t}_2, s, \vec{k}' \end{matrix} \right\}$   
 $\nu_1, \vec{k}$        $\vec{q}, \vec{t}$        $\nu_2, \vec{k}'$

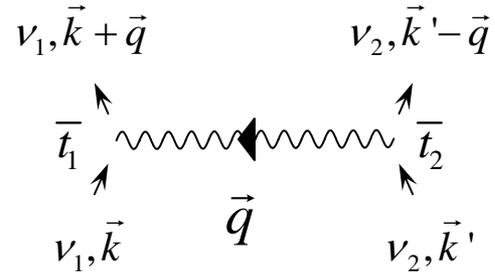
$$\frac{-i}{\hbar} \text{sign}(\nu_1) \frac{i \mathcal{G}^{(0)}(\vec{q}, \vec{t}_1)}{\hbar \nu_1} \frac{1}{\sqrt{2}} \frac{\vec{E}(\vec{q}, \vec{t}_2)}{\sqrt{2}} \delta(\vec{t}_1 - \vec{t}_2)$$



$$\bar{t} \longleftarrow \bar{t}'$$

$$\{v, \vec{k}\}$$

$$iG_{v, \vec{k}}^{(0)}(\bar{t}, \bar{t}')$$



$$\frac{-i}{\hbar} \text{sign}(v_1, v_2) V^c(\vec{q}) \delta(\bar{t}_1 - \bar{t}_2)$$

$$\{h, j, -\vec{k}\} \xleftarrow{\bar{t}} \times \xrightarrow{\bar{t}} \{e, s, \vec{k}\}$$

$$\{h, j, -\vec{k}\} \xrightarrow{\bar{t}} \times \xleftarrow{\bar{t}} \{e, s, \vec{k}\}$$

$$\frac{i}{\hbar} \vec{\mu}_{sj} \cdot \frac{1}{2} \vec{E}(\bar{t})$$

$$\frac{-i}{\hbar} \vec{\mu}_{sj}^* \cdot \frac{1}{2} \vec{E}^*(\bar{t})$$





Next:

More about Green's functions, propagators  
and Feynman diagrams



The idea:

$$\langle \hat{T}_c a_n(t) a_m^\dagger(t') \rangle$$

represent as perturbation series  
via Feynman diagrams

$$\langle \hat{T}_c a_n(t) a_m^\dagger(t') \rangle \Big|_{t'=t+\varepsilon}$$

obtain expectation values  
from equal-time limit



Perturbation theory:  $H = H_0 + H'$

$$\langle \hat{T}_c a_n(t) a_m^\dagger(t') \rangle = \langle \hat{T}_c e^{-\frac{i}{\hbar} \int_c dt'' H_I'(t'')} a_{I,n}(t) a_{I,m}^\dagger(t') \rangle$$

*full Green's function  
(propagators)*

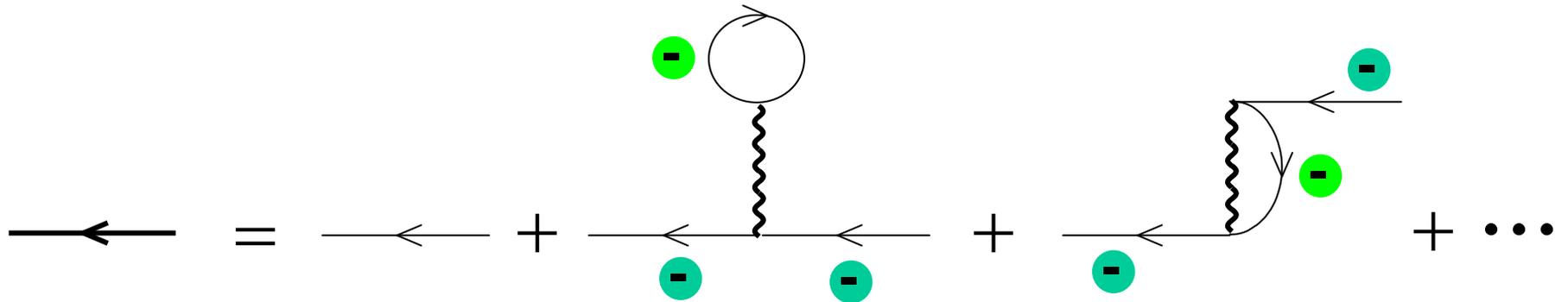
*free particle operators*

Feynman diagrams: expand exponential and factorize:

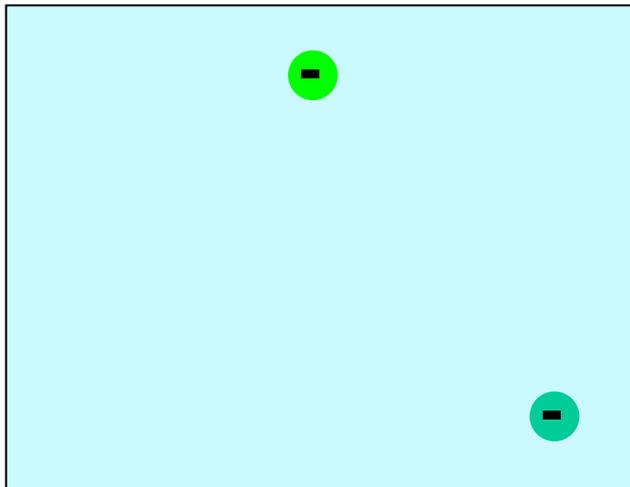
$$e^{ix} = 1 + ix - \frac{1}{2}x^2 + \dots$$

$$\langle a_{I,i} a_{I,j} a_{I,k}^\dagger a_{I,l}^\dagger \rangle = \langle a_{I,i} a_{I,l}^\dagger \rangle \langle a_{I,j} a_{I,k}^\dagger \rangle - \langle a_{I,i} a_{I,k}^\dagger \rangle \langle a_{I,j} a_{I,l}^\dagger \rangle$$

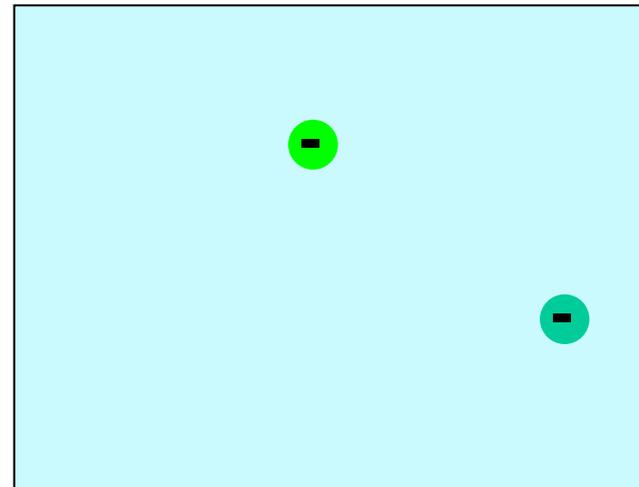




direct Coulomb interaction



exchange Coulomb interaction





## Rules and Regulations (short version)



1. Draw all topologically distinct connected diagrams with two external points
2. Sum over all internal indices
3. Attach an additional factor of  $(-1)$  for each closed Fermion loop

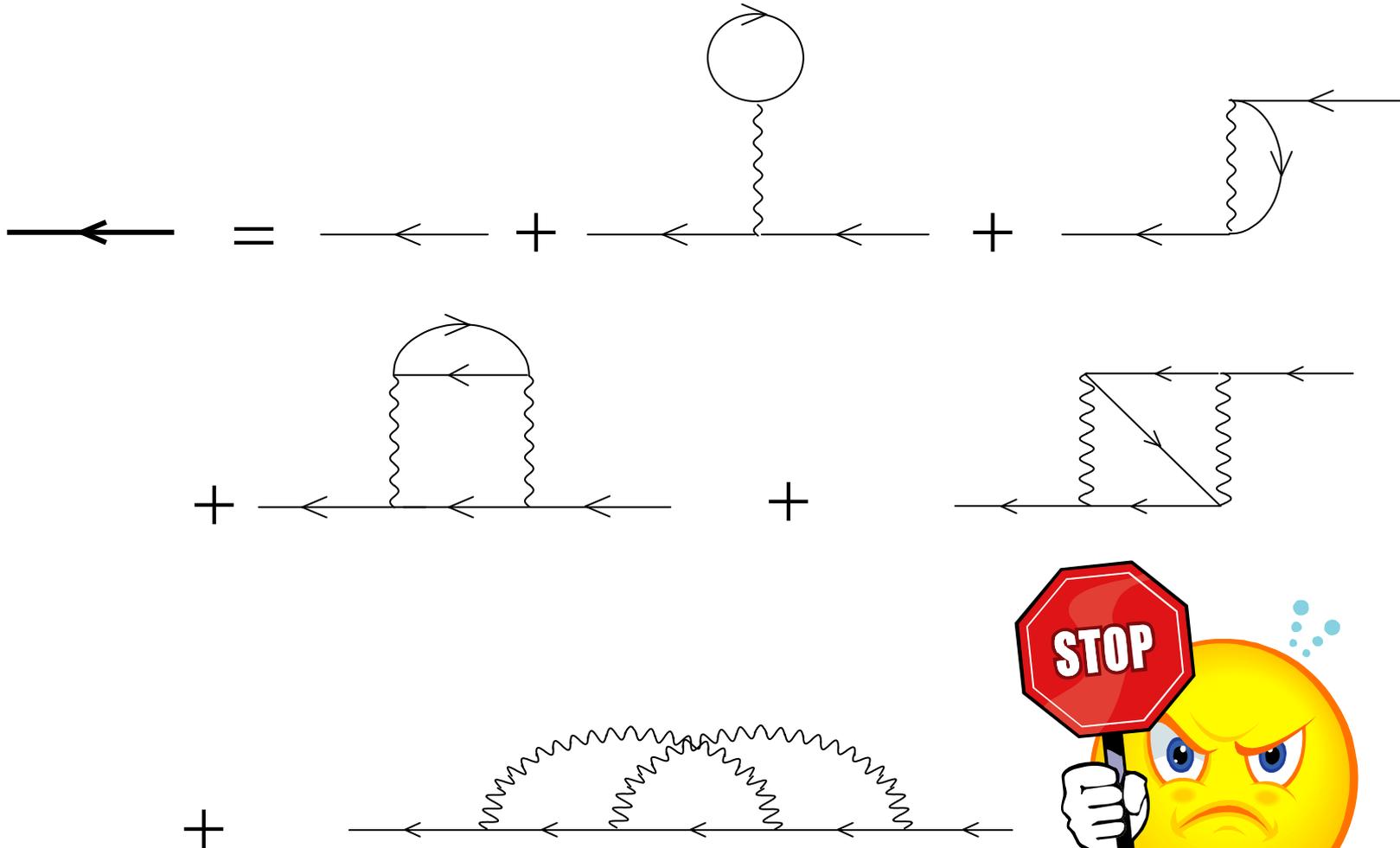
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### Complete rules:

- Thermodynamic equilibrium: Fetter, Walecka, *Quantum Theory of Many Particle Systems*
- Optically excited semiconductors: Kwong, Binder, Phys. Rev. B 61, 8341 (2000)
- Semiconductors with quantized light: Kwong, Rupper, Binder, Phys. Rev. B 79, 155205 (2009)

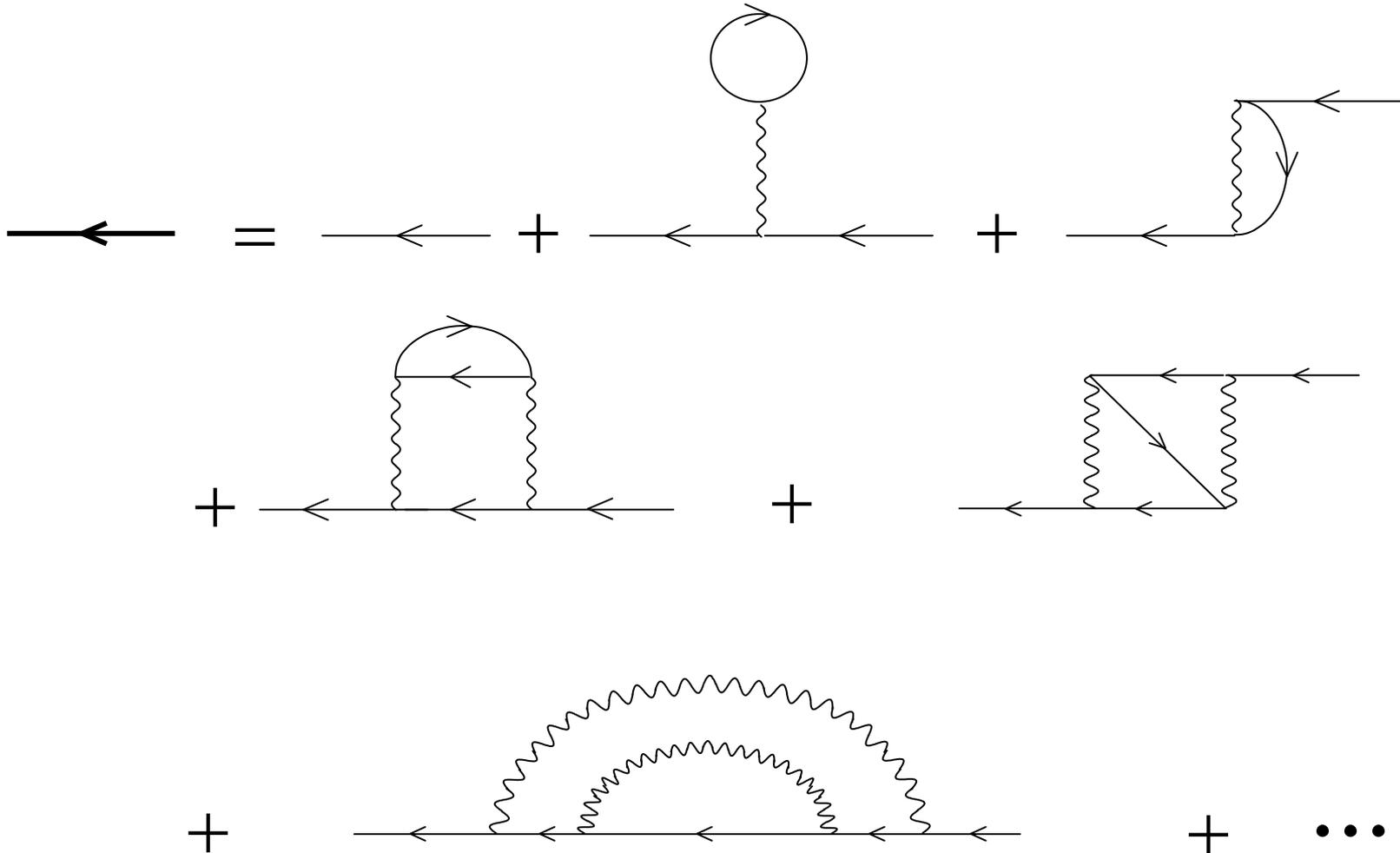


## Wick's theorem: sum up all different graphs





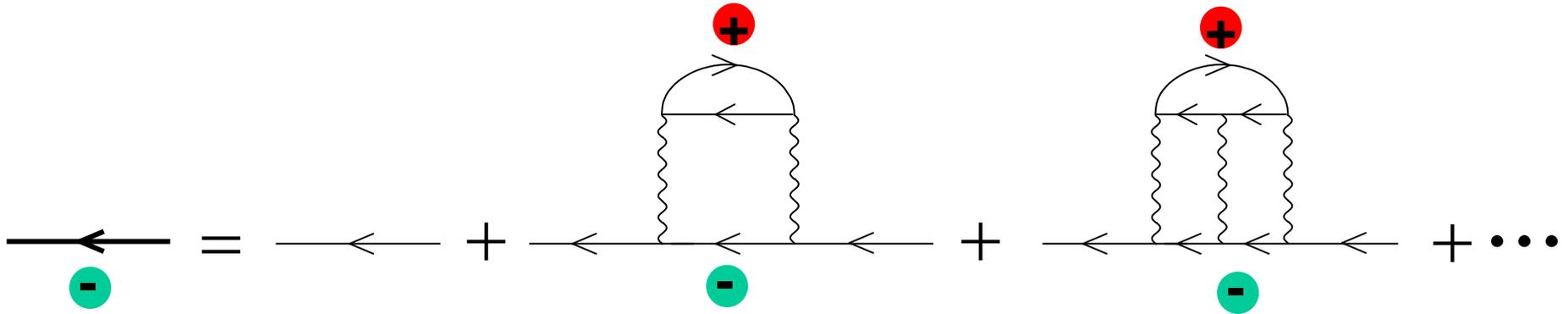
## Wick's theorem: sum up all different graphs

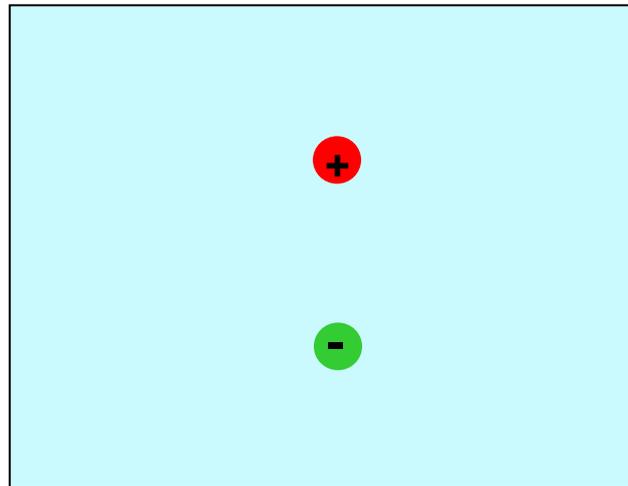
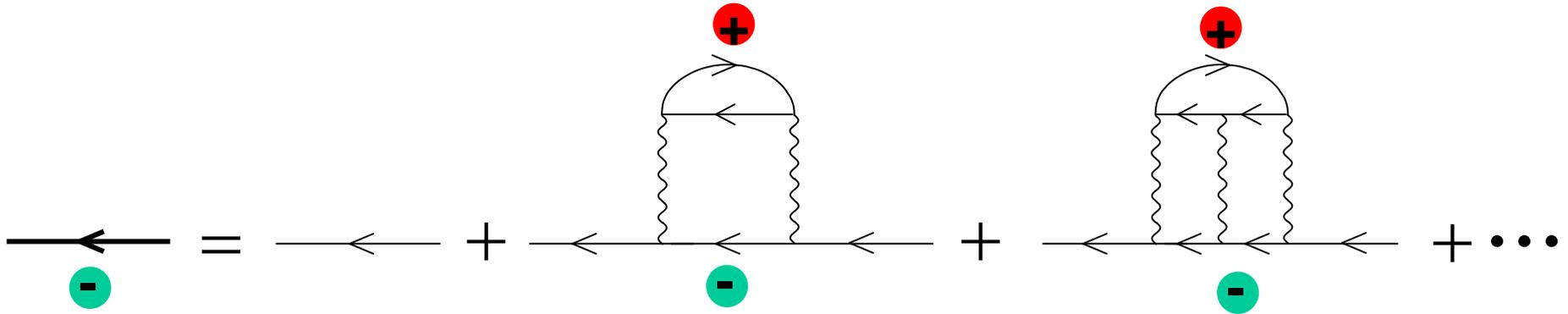






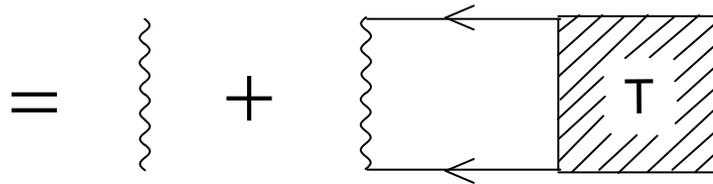
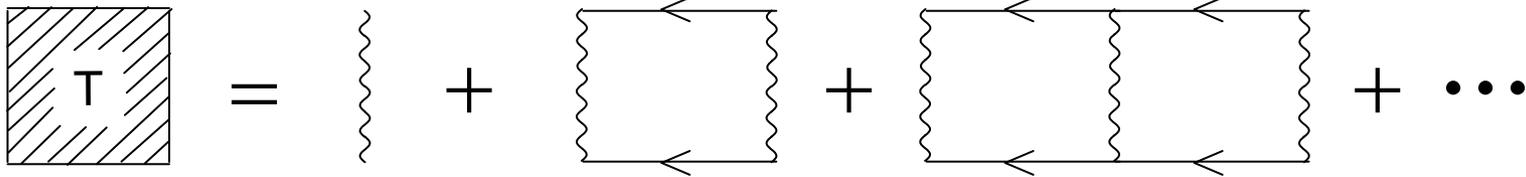
All of them???







## Ladder diagrams



describes non-perturbative Coulomb correlation (including possible **bound states**)

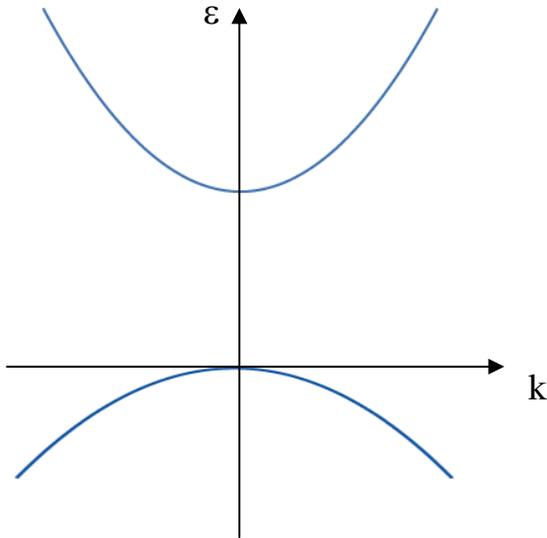


Next:

Third-order nonlinear optical response  
and excitonic diagrams

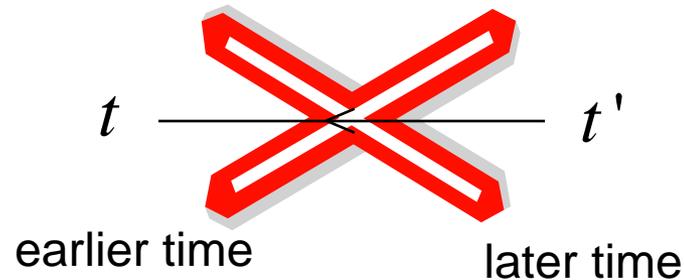


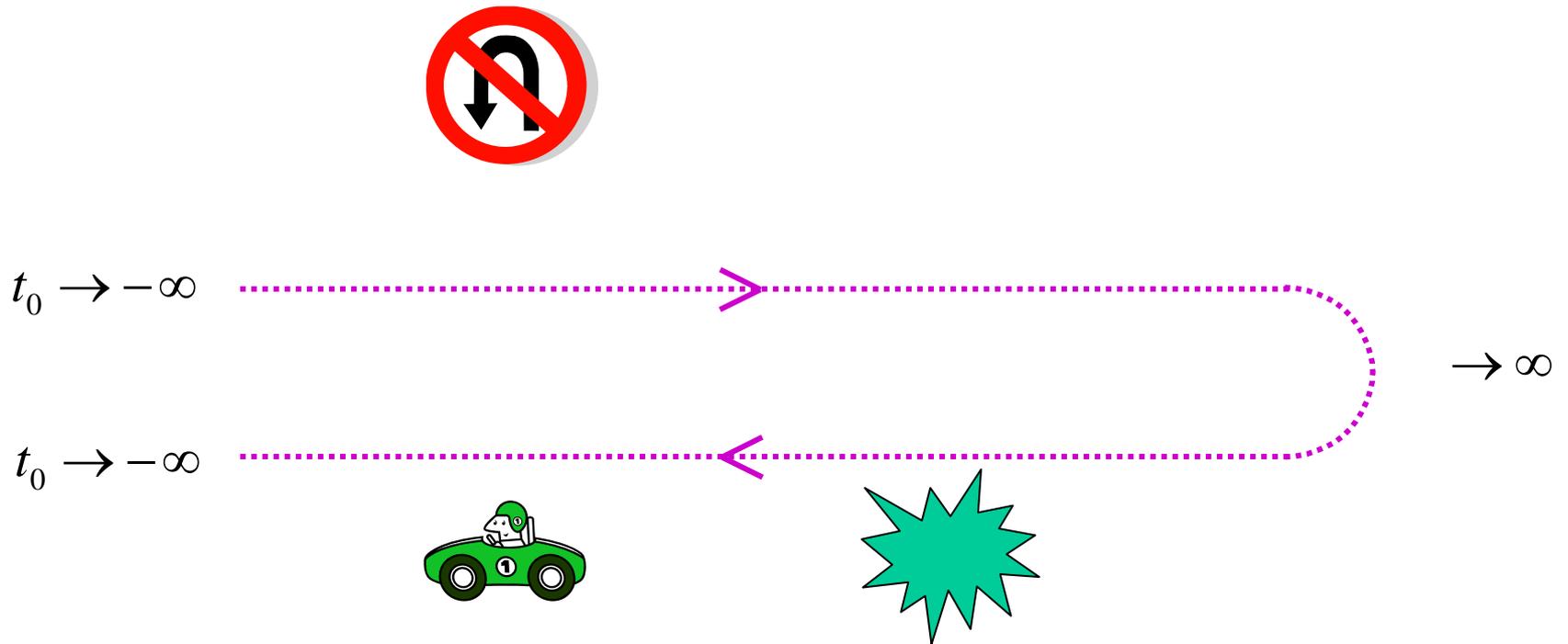
## Dynamics-controlled truncation (DCT) (Axt, Stahl, Z. Phys. B 93, 205 (1994))



Without optical excitation, no density-like propagators:

$$\langle a_I^\dagger(t') a_I(t) \rangle$$



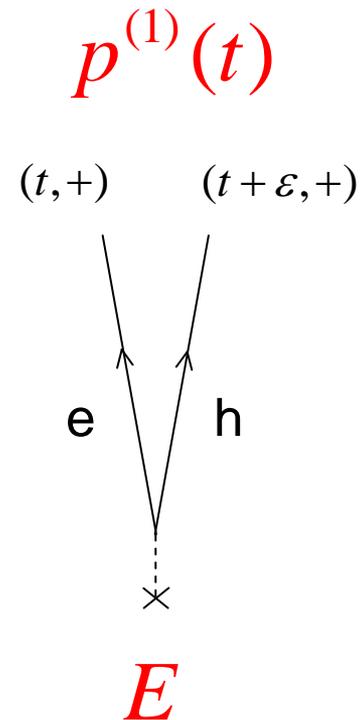


DCT: "Don't Counterpropagate in Time"



## First-order polarization

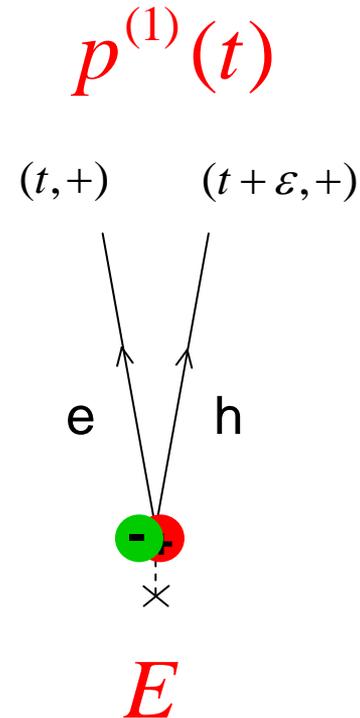
$$\langle a_h(t) a_e(t) \rangle$$





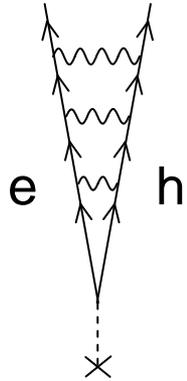
## First-order polarization

$$\langle a_h(t) a_e(t) \rangle$$



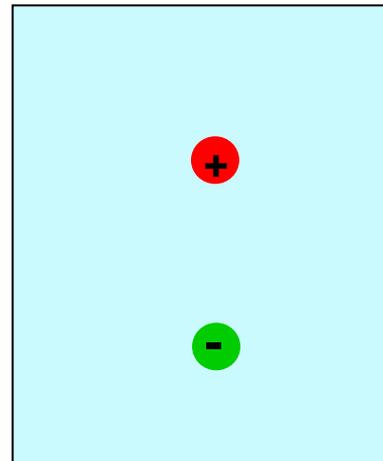
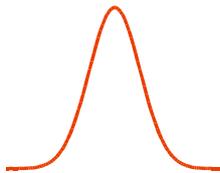


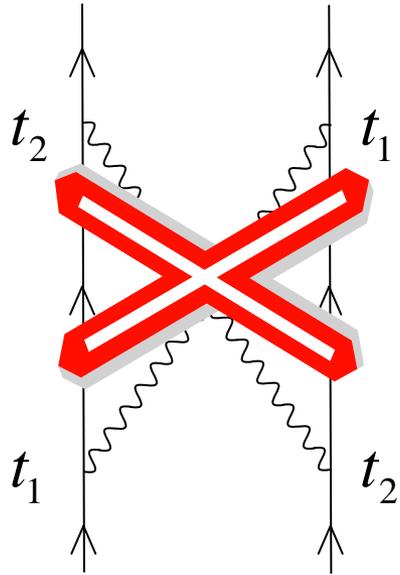
## sum up ladder diagrams: excitons

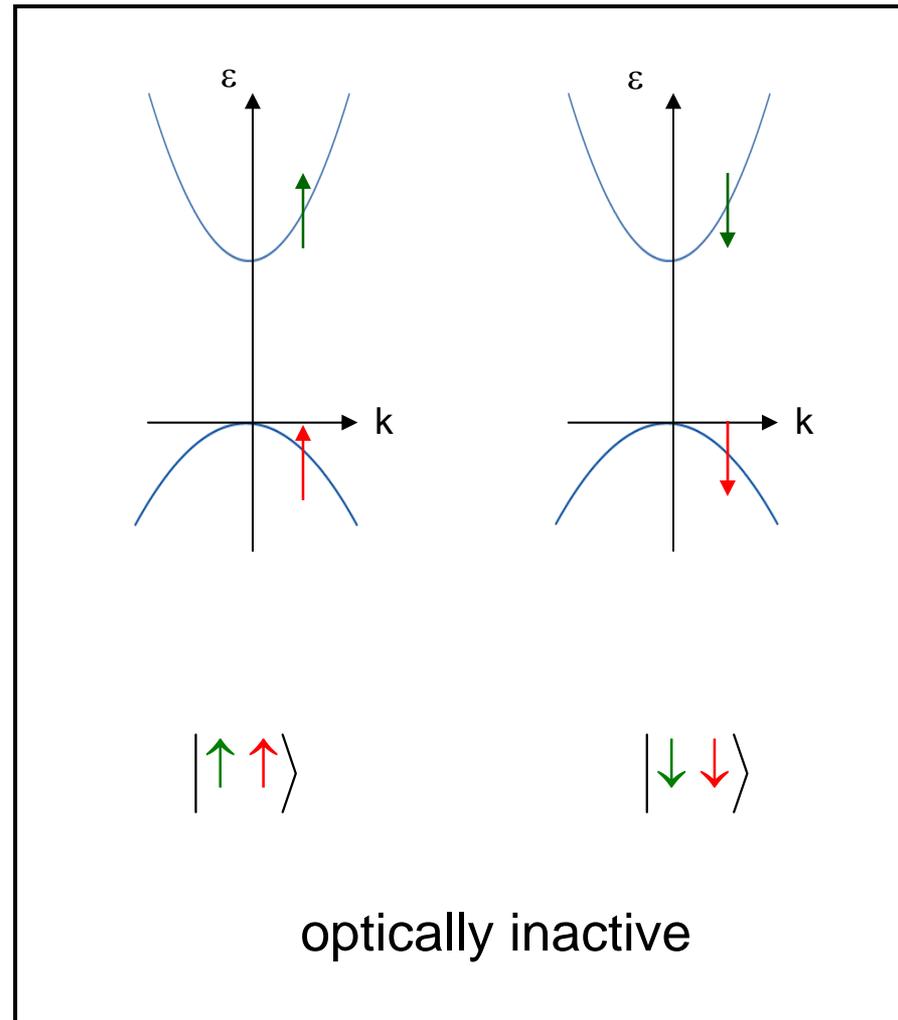
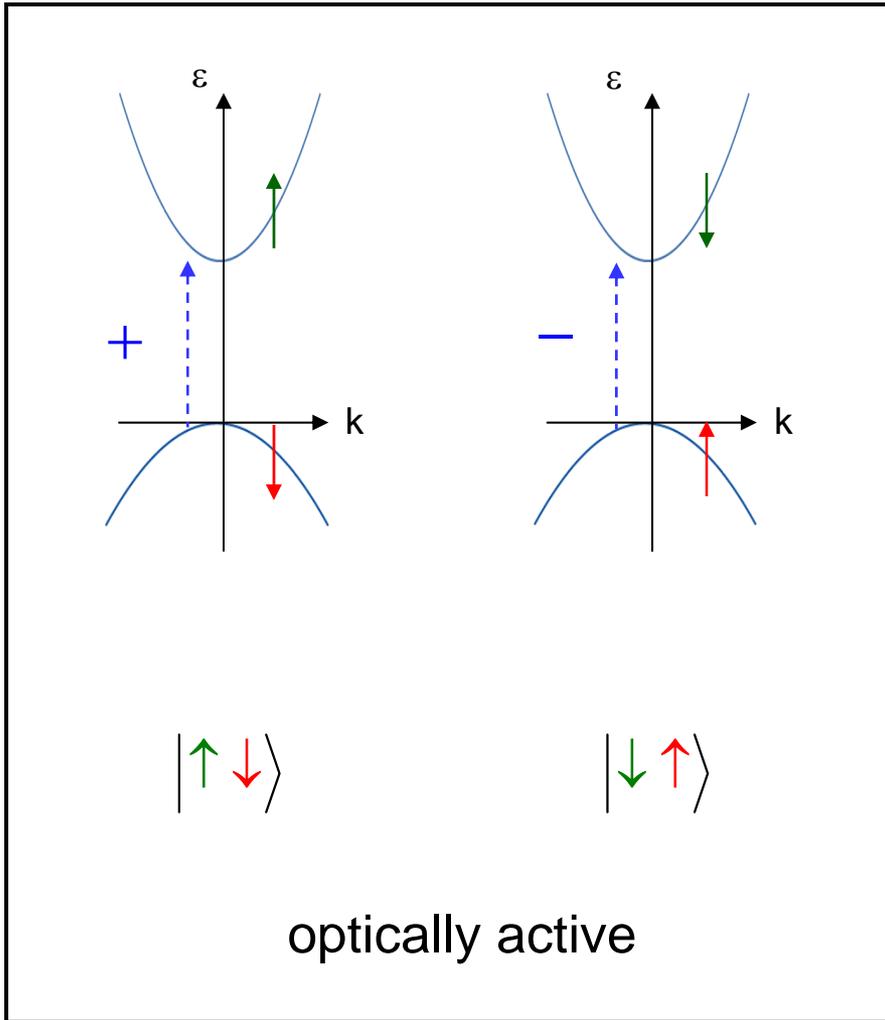


In 1s approximation:  $p_{eh}(\mathbf{k}, \mathbf{q}, t) \cong \phi_{1s}(\mathbf{k}) p(\mathbf{q}, t)$

relative e-h momentum  $\mathbf{k}$       center-of-mass momentum  $\mathbf{q} \cong 0$



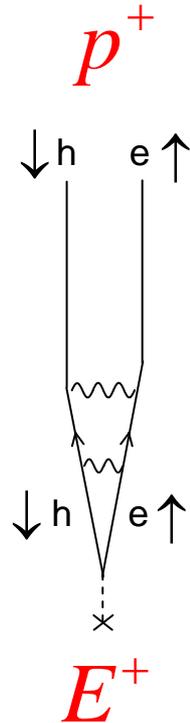




notation: | **electron spin**, **hole spin**  $\rangle$



## First order excitonic response



In 1s approximation:

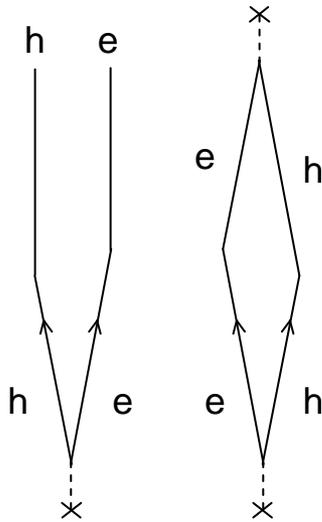
$$i\hbar \dot{p}^+ = (\epsilon_x - i\gamma) p^+ - \phi^*(0) E^+$$

with  $E^+ \equiv d_{cv} E^+$



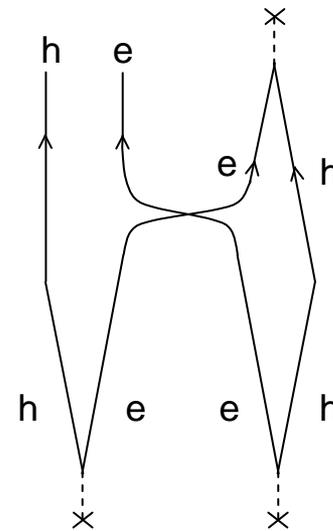
## The two basic third-order diagrams

$$\langle a_h a_e \rangle$$



*direct*

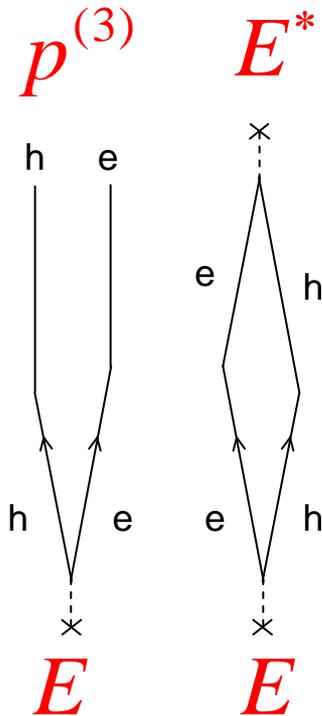
$$\langle a_h a_e \rangle$$



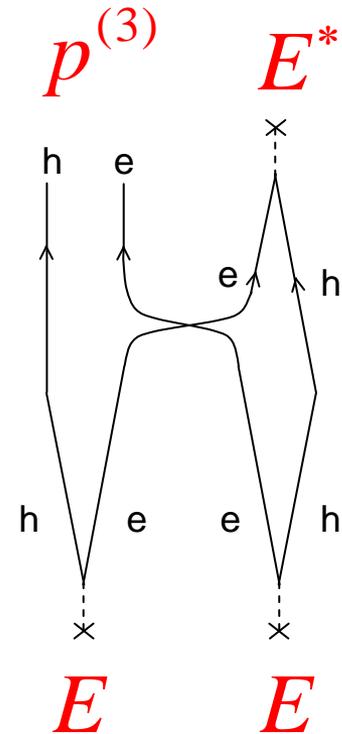
*electron exchange*



## The two basic third-order diagrams



*direct*

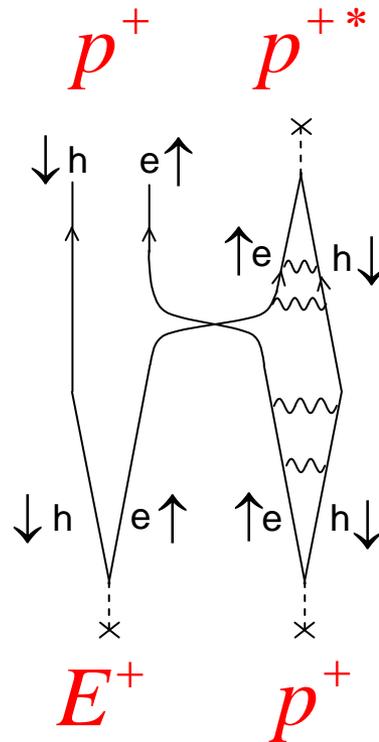


*electron exchange*

Without Coulomb interaction,  
this diagram is disconnected  
(does not contribute)



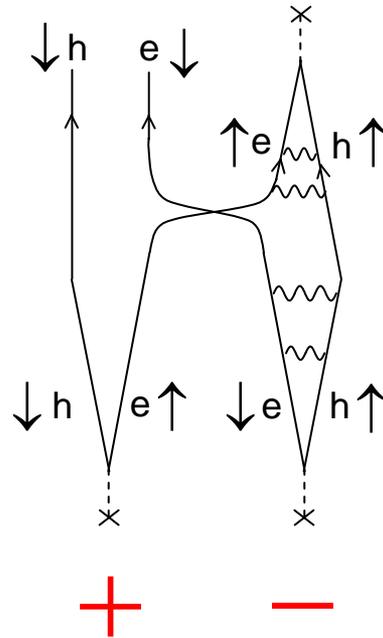
## Phase-Space Filling (PSF)



$$i\hbar \dot{p}^+ = (\epsilon_x - i\gamma) p^+ - \phi_{1s}^*(0) E^+ + 2A^{\text{PSF}} |p^+|^2 E^+$$



not possible:



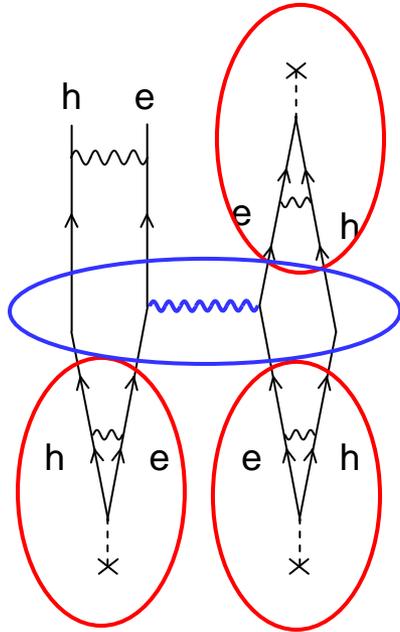
$|\downarrow\downarrow\rangle$  and  $|\uparrow\uparrow\rangle$

not optically active

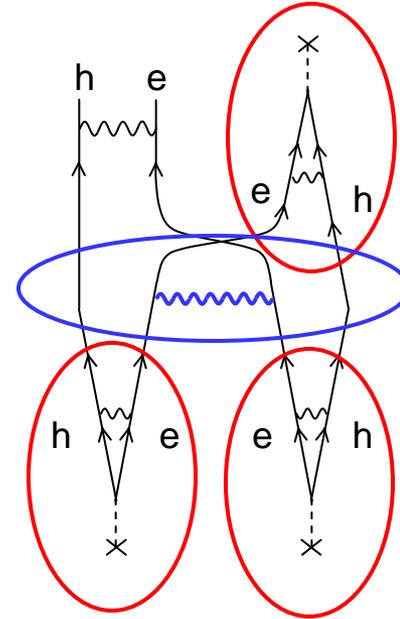
$$A^{\text{PSF}} p^+ p^- E^+$$



"direct"

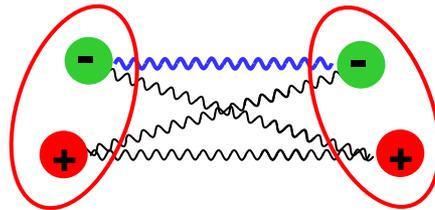


*exciton-exciton  
interaction  
(1 of 4 contributions)*



"exchange"

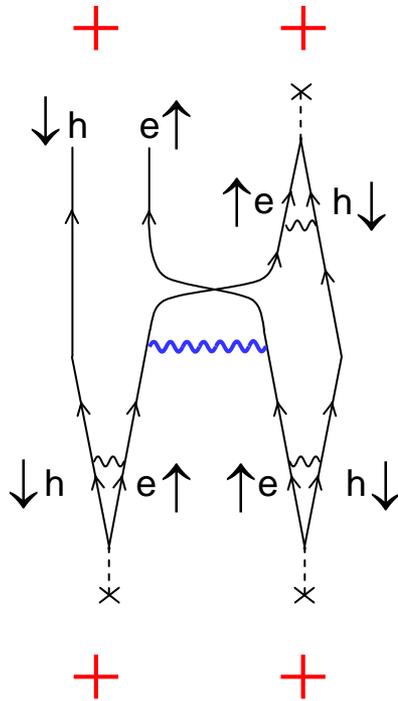
*excitons  
 $q \cong 0$*



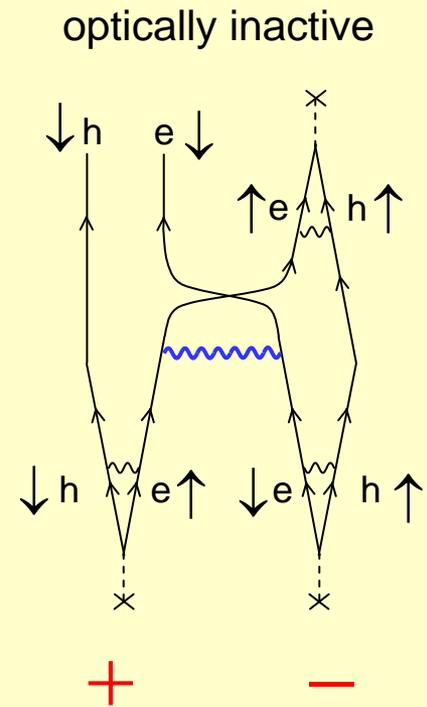
$$i\dot{p}|_{HF} = (W^{dir} + W^{exc}) |p|^2 p$$



$$i\hbar \dot{p}^+ = (\epsilon_x - i\gamma) p^+ - (\text{PSF term}) + V^{\text{HF}} |p^+|^2 p^+$$



Hartree term zero, Fock term only ++



cannot be:  $|p^+|^2 p^-$



## Interacting Electron Theory of Coherent Nonlinear Response

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(Received 11 July 1994)

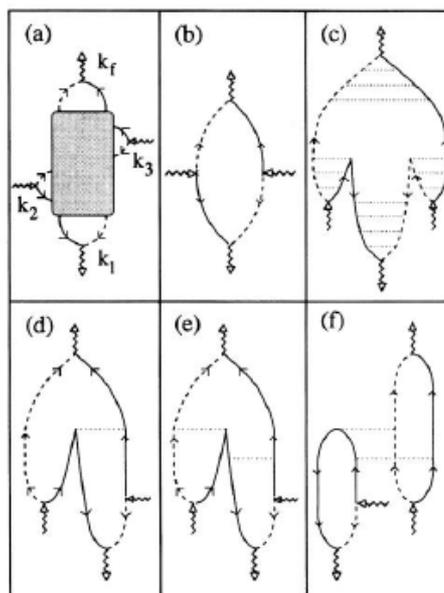
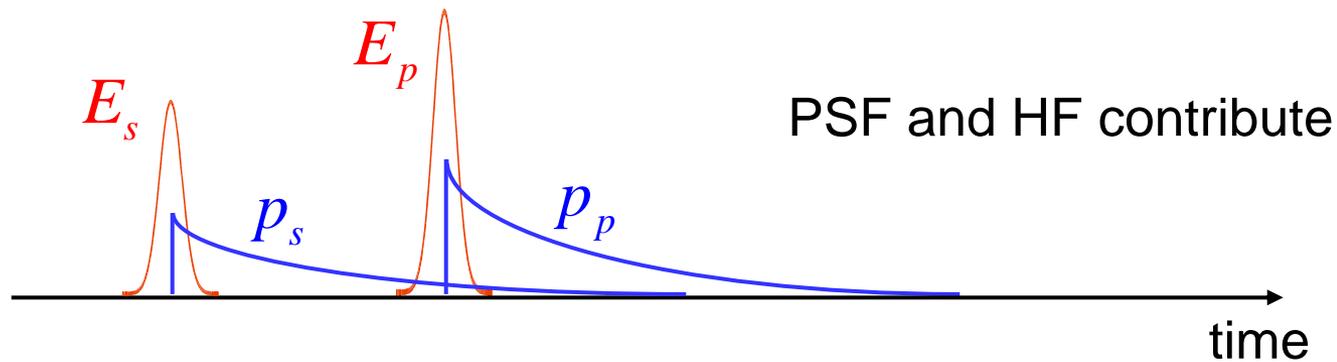


FIG. 1. Diagrammatic representation for the  $\chi_3$  processes. For explanation, see text.

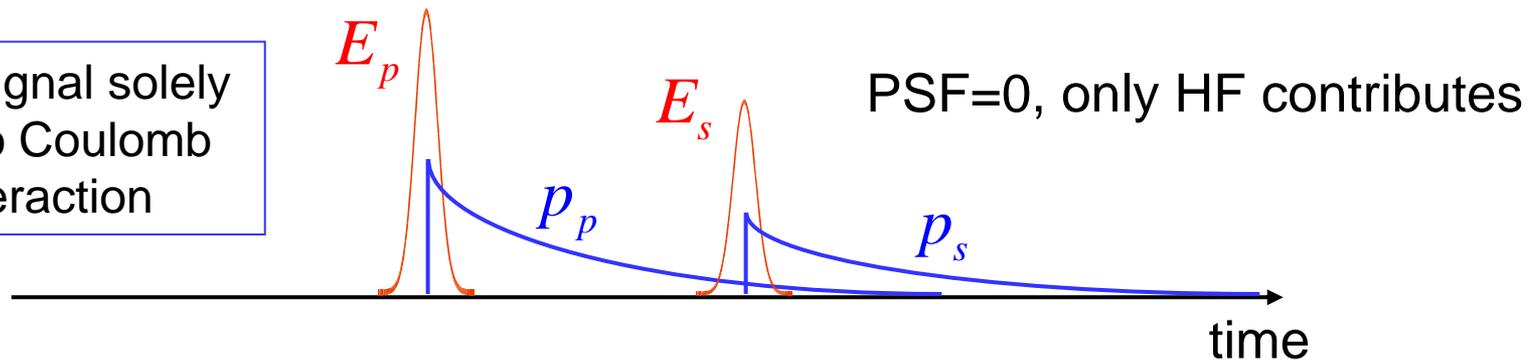


## Phase-space filling vs Hartree-Fock in FWM

$$i\hbar \dot{p}_f = (\varepsilon_x - i\gamma) p_f + 2A^{\text{PSF}} p_s^* p_p E_p + V^{\text{HF}} p_s^* p_p p_p$$



FWM signal solely due to Coulomb interaction





## Line shape of time-resolved four-wave mixing

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*AT&T Bell Laboratories, Holmdel, New Jersey 07733*

S. Schmitt-Rink  
*AT&T Bell Laboratories, Murray Hill, New Jersey 07974*

W. Schäfer  
*Forschungszentrum Jülich, Hochleistungsrechenzentrum, 5170 Jülich, Federal Republic of Germany*  
(Received 8 May 1990)

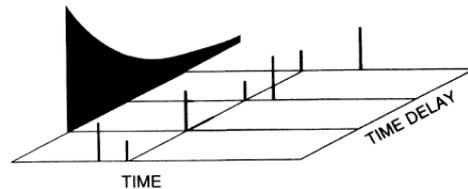


FIG. 5. Graphical representation of the predictions of the noninteracting two-level model of Ref. 3 for  $\delta$  pulses. The temporal position of the two incident pulses is marked by the spikes. Pulse 2 is displayed with twice the strength of pulse 1. On the second line the two pulses overlap, which corresponds to zero time delay. The light areas represent the square of the magnitude of the third-order polarization as a function of time for various time delays. The full area on the left is the energy of the diffracted signal as a function of the time delay (i.e., the time integral of the light area at a given time delay).

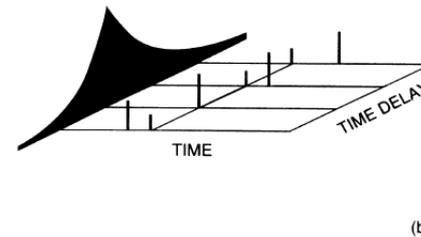


FIG. 7. Same as Fig. 5, but including local-field corrections. Results for (a) a homogeneously broadened line and (b) a strongly inhomogeneously broadened line are depicted. In contrast to the noninteracting two-level model, Fig. 5, one also finds a signal for negative time delay. This additional signal, however, is quite sensitive to inhomogeneous broadening and eventually disappears completely for a strongly inhomogeneously broadened line.

homogeneously  
broadened

inhomogeneously  
broadened



### Effects of Coherent Polarization Interactions on Time-Resolved Degenerate Four-Wave Mixing

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S. Schmitt-Rink  
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W. Schäfer  
*Forschungszentrum Jülich, Höchstleistungsrechenzentrum, D-5170 Jülich, Federal Republic of Germany*  
(Received 13 April 1990)

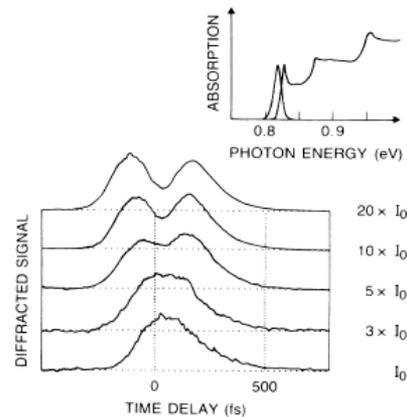


FIG. 3. DFWM signals in InGaAs/InAlAs vs time delay for a lattice temperature of 5 K, 10-meV laser detuning, and different excitation intensities. Inset: Sample absorption at 5 K and the pulse spectrum.

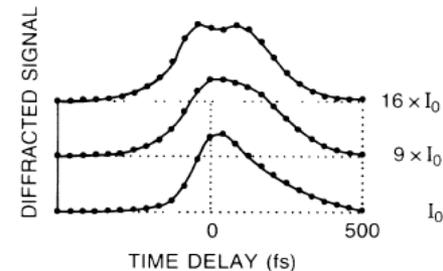
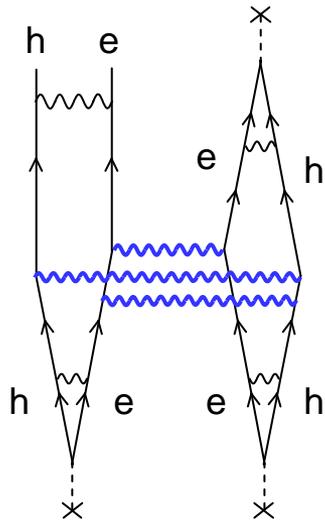


FIG. 4. Calculated DFWM signals vs time delay for different excitation intensities.



## Beyond Hartree-Fock: Excitonic Correlation Functions



Excitonic correlation functions:

$$G^{++} \leftrightarrow \text{sum} (W^{dir} - W^{exc})$$

$$G^{+-} \leftrightarrow \text{sum} (W^{dir} - W^{exc})$$

$$+ \text{sum} (W^{dir} + W^{exc})$$



$$i\hbar \dot{p}^+ = (\varepsilon_x - i\gamma) p^+ - [\phi_{1s}^*(0) - 2A^{\text{PSF}} |p^+|^2] E^+$$

Phase-space filling

$$+V^{\text{HF}} |p^+|^2 p^+$$

Hartree-Fock  
Coulomb interaction

$$+2 p^{+*} \int_{-\infty}^{\infty} dt' G^{++}(t-t') p^+(t') p^+(t')$$

$$+ p^{-*} \int_{-\infty}^{\infty} dt' G^{+-}(t-t') p^+(t') p^-(t')$$

} Time-retarded  
two-exciton  
correlations  
(incl. biexciton)



Same equation for the coherent third order interband polarization:

- **Dynamics Controlled Truncation**  
Axt , Stahl, Z. Phys. B 93, 195 (1994)
- **Hubbard operators, force-force correlation function**  
Oestreich, Schoenhammer, Sham, Phys. Rev. B 58,12920 (1998)
- **Cumulant expansions**  
Meier, Koch, Phys. Rev. B 59, 13202 (1999);  
Hoyer, Kira, S.W. Koch, Phys. Rev. B 67, 155113 (2003)
- **Nonequilibrium Green's functions**  
Kwong, Binder, Phys. Rev. B 61, 8341 (2000)

see also: Schäfer, Wegener, *Semiconductor Optics and Transport Phenomena*  
(Springer, Berlin, 2002)



## Degenerate FWM (all fields at frequency $\omega$ )

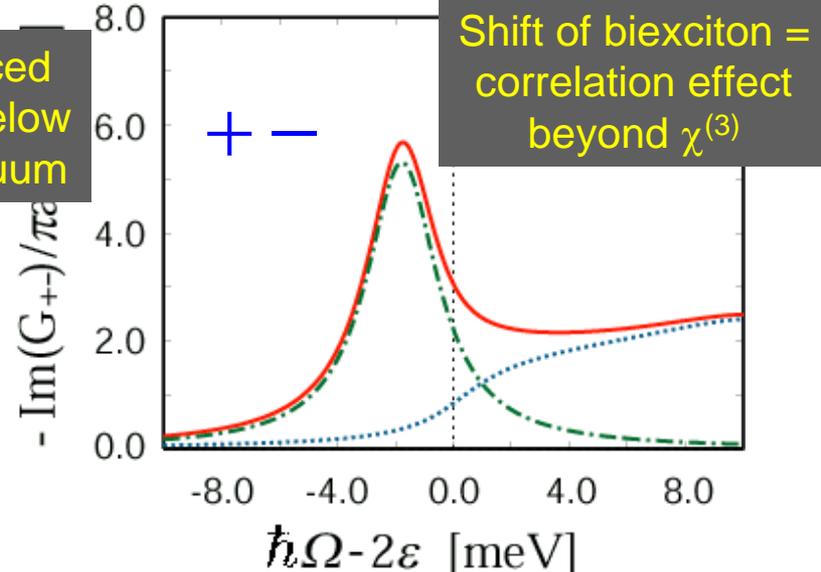
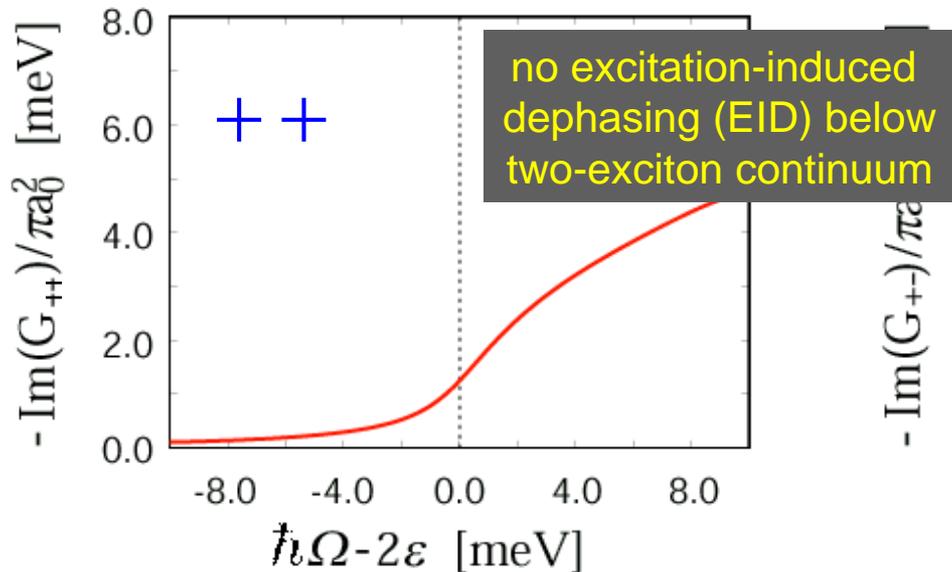
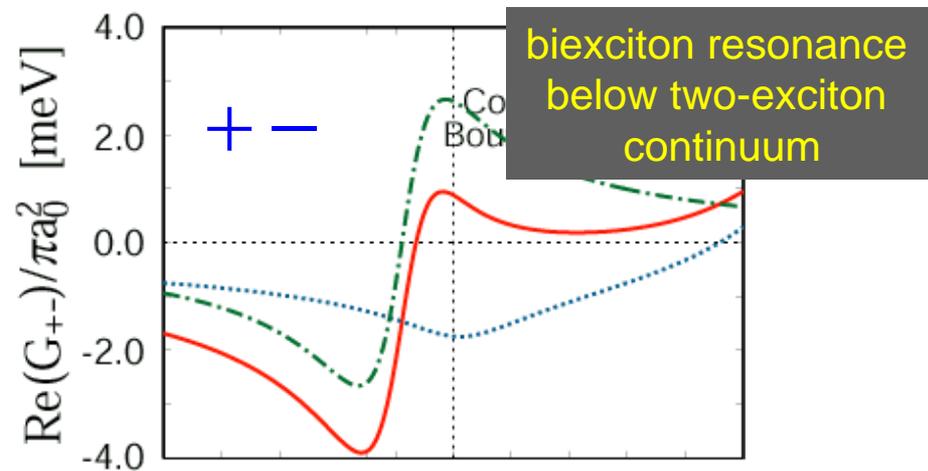
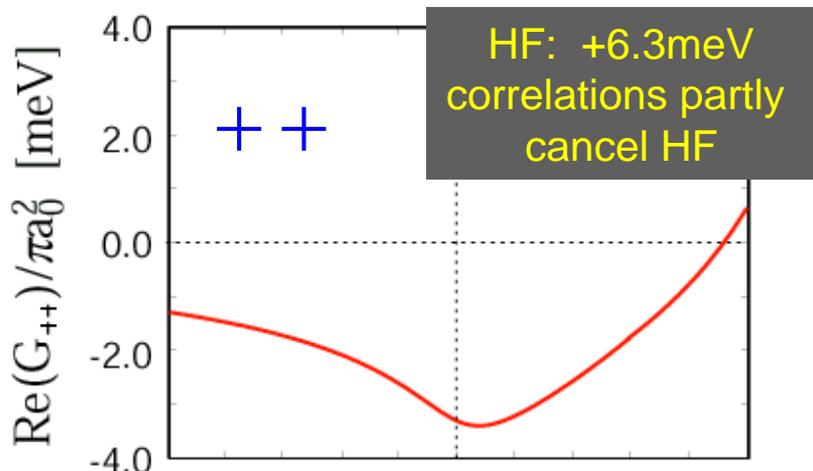
$$p_f^\pm(\omega) = \chi^{++}(\omega) E_{p\pm}^2(\omega) E_{s\pm}^*(\omega) + \chi^{+-}(\omega) E_{s\mp}^*(\omega) E_{p\pm}(\omega) E_{p\mp}(\omega)$$

$$\chi^{++}(\omega) \sim \left| \chi^{(1)}(\omega) \right|^2 \left[ \chi^{(1)}(\omega) \right] \left\{ G^{PSF}(\omega) + V^{HF} + 2G^{++}(2\omega) \right\}$$

$$\chi^{+-}(\omega) \sim \left| \chi^{(1)}(\omega) \right|^2 \left[ \chi^{(1)}(\omega) \right] G^{+-}(2\omega)$$

$$\text{with } \chi^{(1)}(\omega) \sim \frac{1}{\omega - \varepsilon_x + i\gamma} \quad G^{PSF}(\omega) \sim 1 / \chi^{(1)}(\omega)$$

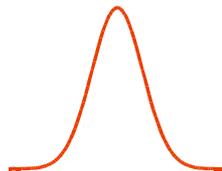
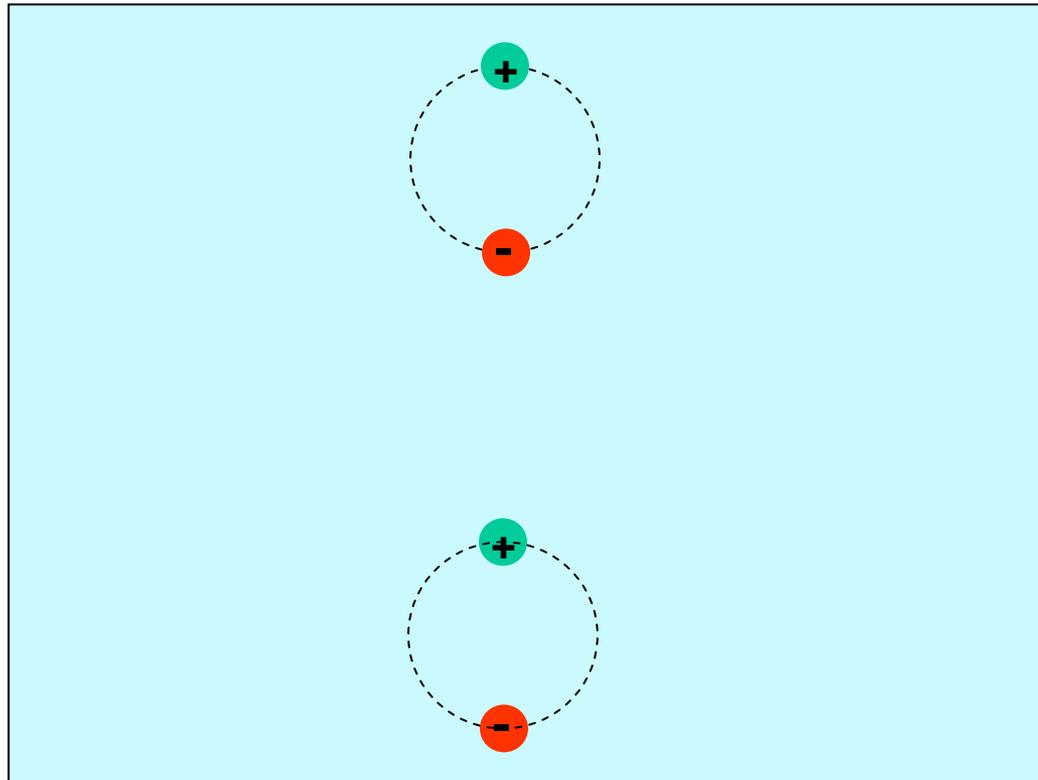
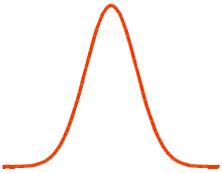
- Takayama, Kwong, Romyantsev, Kuwata-Gonokami, Binder, JOSA-B 21, 2164 (2004)
- Kwong, Takayama, Romyantsev, Kuwata-Gonokami, Binder, Phys. Rev. B 64, 045316 (2001)





$$G^{+-} p^+ p^-$$

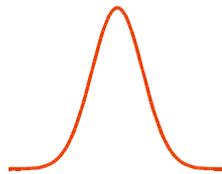
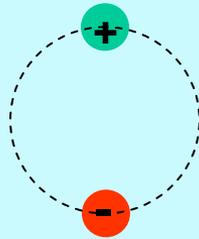
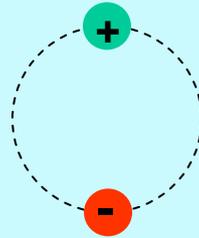
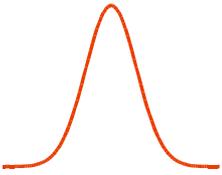
includes bound two-exciton states (biexciton)

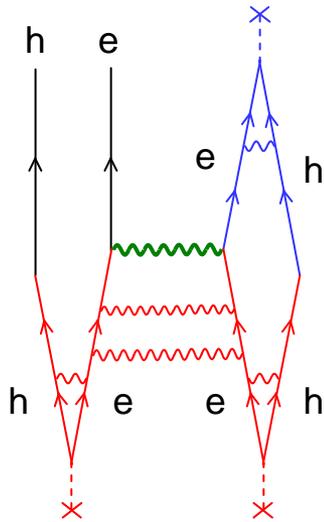




$$G^{++} p^+ p^+$$

only continuum states





$$i \frac{d}{dt} \langle a a \rangle \Big|_{corr} = W \langle a^\dagger a^\dagger \rangle \langle a a a a \rangle_{corr}$$

Hierarchy of correlation functions

$$\langle a a \rangle \xleftrightarrow{W} \langle a a a a \rangle \xleftrightarrow{W} \langle a a a a a a \rangle \xleftrightarrow{W} \dots$$



## Hierarchy of correlation functions

$$\langle \underbrace{a_1^+ \cdots a_n^+}_n \underbrace{a_1 \cdots a_m}_m \rangle$$

### Proven:

If initial e-h density zero, one has, exact to order of  $E$ ,

- truncation of hierarchy
- factorization to yield closed set of equations of motion

Axt, Stahl, Z. Phys. B 93, 205 (1994)

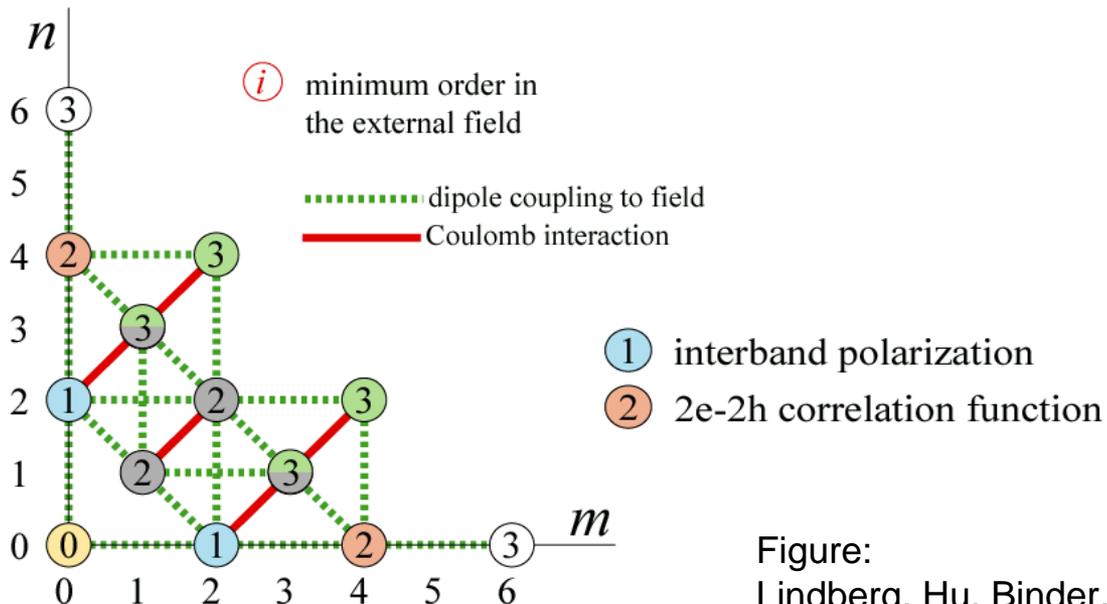


Figure:

Lindberg, Hu, Binder, Koch, Phys. Rev. B 50, 18060 (1994)



Next:

Some experimental FWM data



# Time-resolved measurements of the polarization state of four-wave mixing signals from GaAs multiple quantum wells

A. E. Paul, J. A. Bolger, and Arthur L. Smirl

Laboratory for Photonics and Quantum Electronics, 100 Iowa Advanced Technology Laboratories, University of Iowa, Iowa City, Iowa 52242-1000

J. G. Pellegrino

National Institute of Standards and Technology, Gaithersburg, Maryland 20899

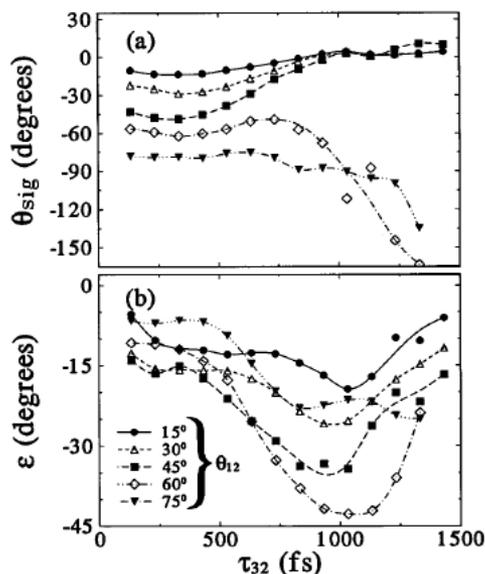
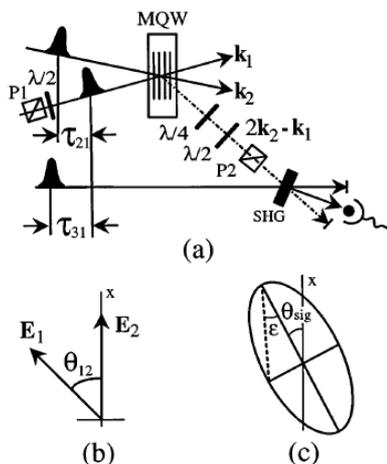


Fig. 2. Measurements of (a) the azimuthal angle  $\theta_{sig}$  and (b) the ellipticity angle  $\epsilon$  as a function of time for selected angles  $\theta_{12}$  between the two input polarizations for a total peak fluence of  $1.0 \mu\text{J}/\text{cm}^2$ . The curves connect the data points only as a rough guide for the eye.

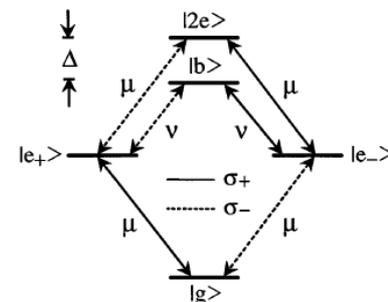
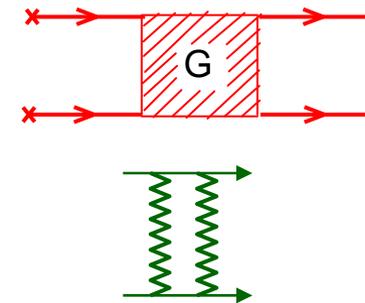
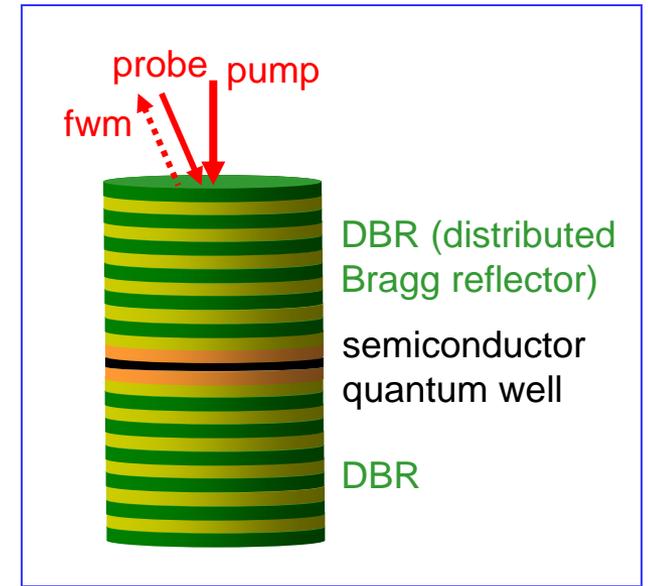
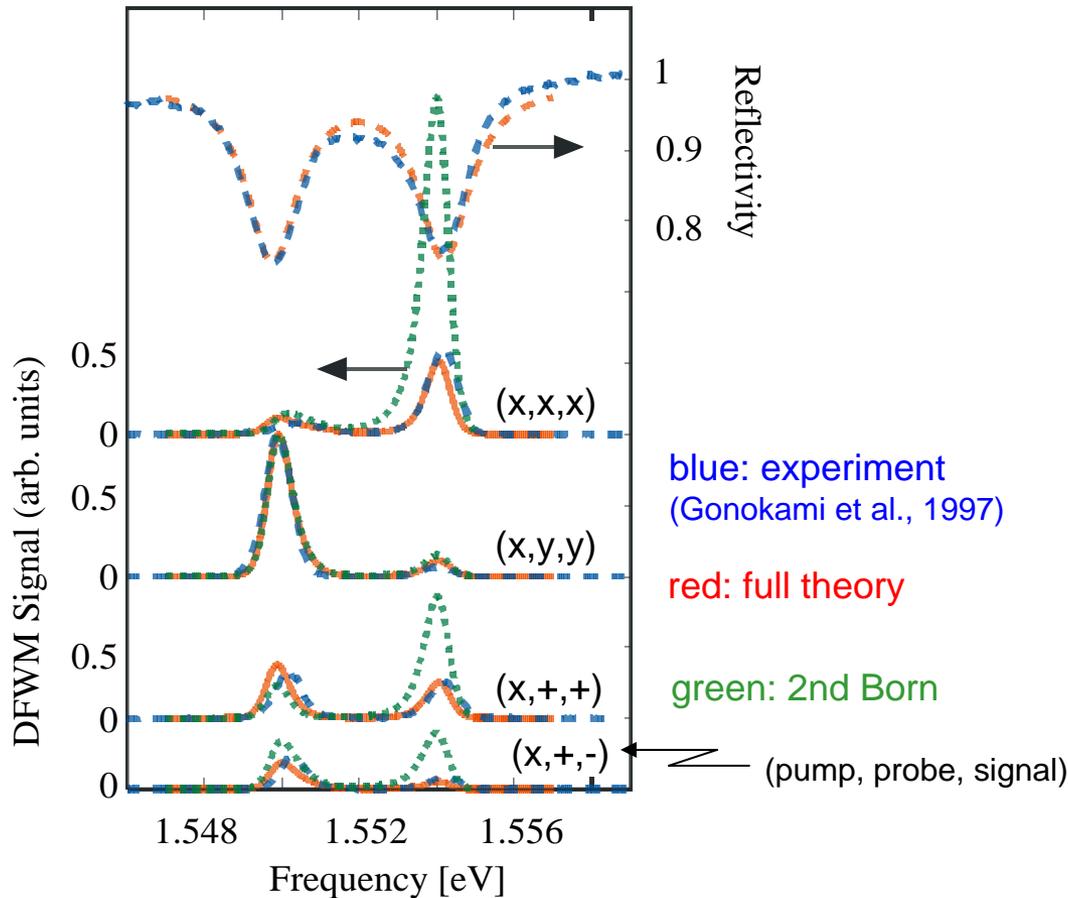


Fig. 6. Schematic of the five-level two-particle system describing the ground state  $|g\rangle$ , the single-exciton states  $|e_{\pm}\rangle$ , the biexciton state  $|b\rangle$ , and the unbound two-exciton state  $|2e\rangle$ . The solid (dashed) lines represent transitions coupled by  $\sigma_{+}$  ( $\sigma_{-}$ ) light,  $\hbar\Delta$  is the biexciton binding energy, and  $\mu$  and  $\nu$  correspond to the exciton and the biexciton dipole matrix elements, respectively.

Identified biexciton, "local field" (HF), and EID



## Signature of non-perturbative continuum correlations



- Kwong, Takayama, Romyantsev, Kuwata-Gonokami, Binder, Phys. Rev. Lett. 87, 27402 (2001)
- Kuwata-Gonokami, Inoue, Suzuura, Shirane, Shimano, Phys. Rev. Lett. 79, 1341 (1997)



## Coherent Dynamics of Excitonic Nonlinear Optical Response in the Nonperturbative Regime

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(Received 24 June 1998)

### microcavity FWM

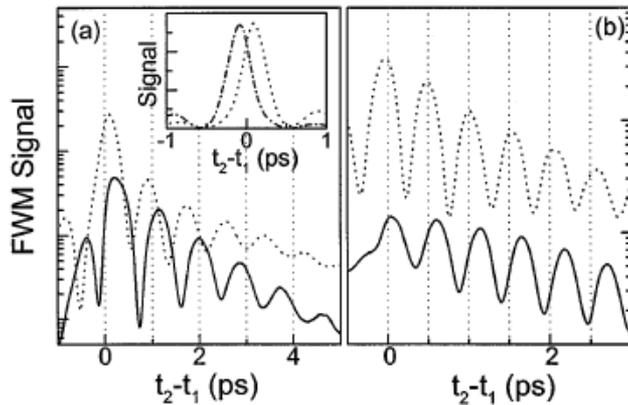


FIG. 1. Time-integrated FWM response for the upper (solid lines) and lower (dashed lines) cavity-polariton modes. (a) At zero exciton cavity detuning. (b) The cavity mode is 3 nm below the hh exciton line center. The inset in (a) also shows FWM responses near  $t_2 - t_1 = 0$  along the directions of  $2\mathbf{k}_2 - \mathbf{k}_1$  (dashed lines) and  $2\mathbf{k}_1 - \mathbf{k}_2$  (dotted lines) for the lower cavity-polariton mode.

Experiment

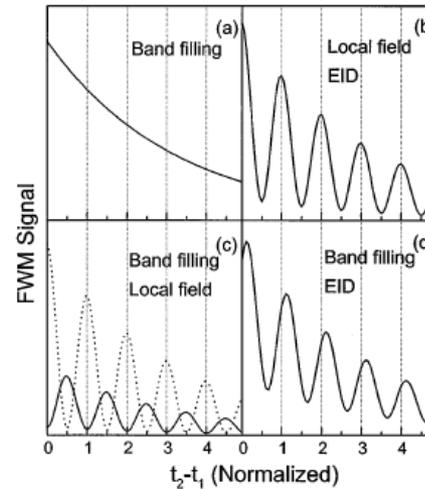


FIG. 3. Calculated time-integrated FWM response due to mechanisms indicated in the figure, where we have assumed  $\delta = 0$ ,  $\epsilon/\Omega = -0.5$ ,  $\sigma N_0/\Omega = 0.8$ ,  $\gamma/\Omega = 0.05$ , and  $\Omega = 2.5$  meV. The time delay is normalized to the oscillation period. Except for (c), FWM responses are identical for both cavity-polariton modes. For (c), the solid (dashed) line is for the upper (lower) cavity-polariton mode.

identified importance  
of EID and local field

Theory



## Quantum Interference of Virtual and Real Amplitudes in a Semiconductor Exciton System

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(Received 7 March 2002; published 18 November 2002)

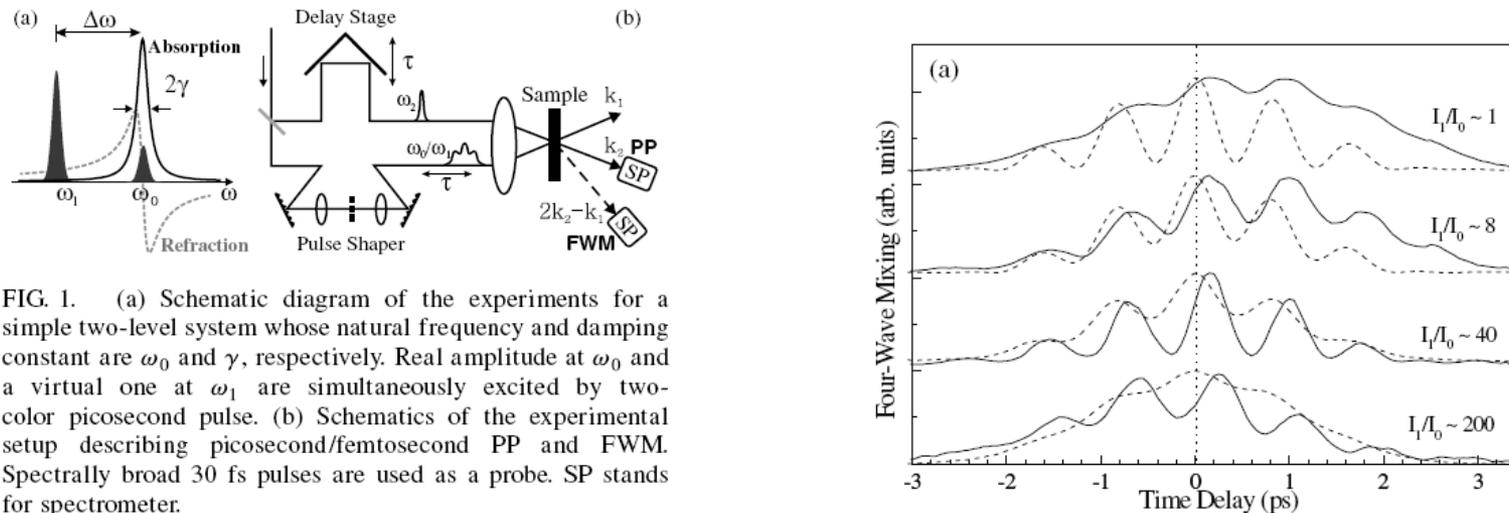


FIG. 1. (a) Schematic diagram of the experiments for a simple two-level system whose natural frequency and damping constant are  $\omega_0$  and  $\gamma$ , respectively. Real amplitude at  $\omega_0$  and a virtual one at  $\omega_1$  are simultaneously excited by two-color picosecond pulse. (b) Schematics of the experimental setup describing picosecond/femtosecond PP and FWM. Spectrally broad 30 fs pulses are used as a probe. SP stands for spectrometer.

two-color pump  
(at and below exciton)

$$\Rightarrow G(\omega_1 + \omega_2)$$

90° phase shift between FWM  
and cross correlation



*Next:*

Correlations beyond  $\chi^{(3)}$



$$i\dot{p} =$$

$$V p^* p p$$

$$p^* [G p p]$$

---

$$V n p$$

$$[G n p]$$

$$V \langle a^\dagger a^\dagger \rangle \langle a a \rangle \langle a a \rangle$$

$$V \langle a^\dagger a^\dagger \rangle \langle a a a a \rangle_{corr}$$

$$V \langle a^\dagger a^\dagger a a \rangle_{corr} \langle a a \rangle$$

$$V \langle a^\dagger a^\dagger a a a a \rangle_{np}$$

HF

biexciton

"incoh. density"

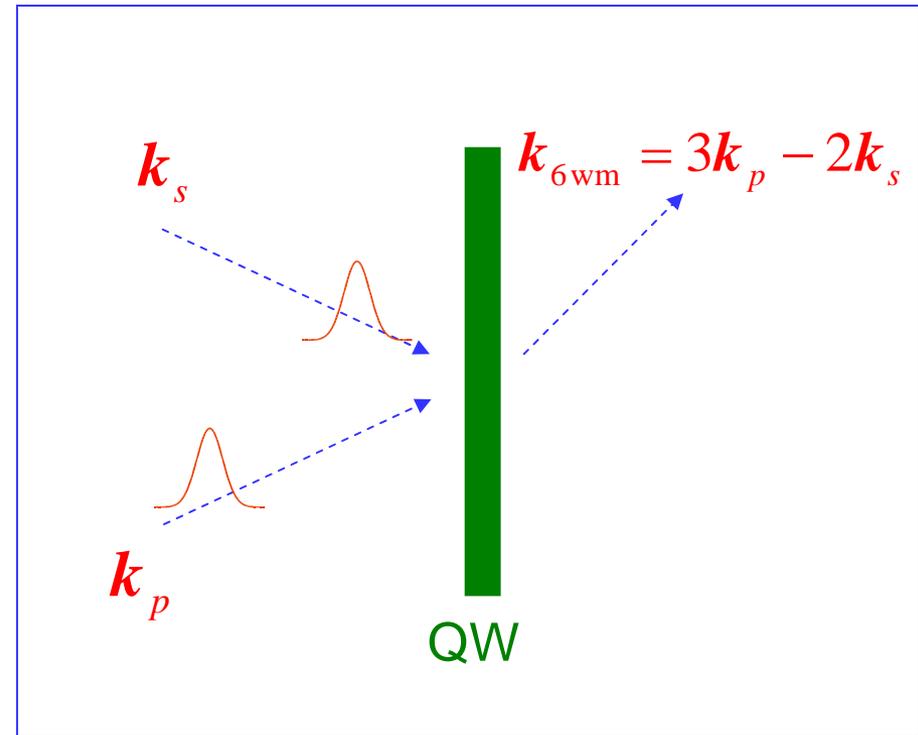
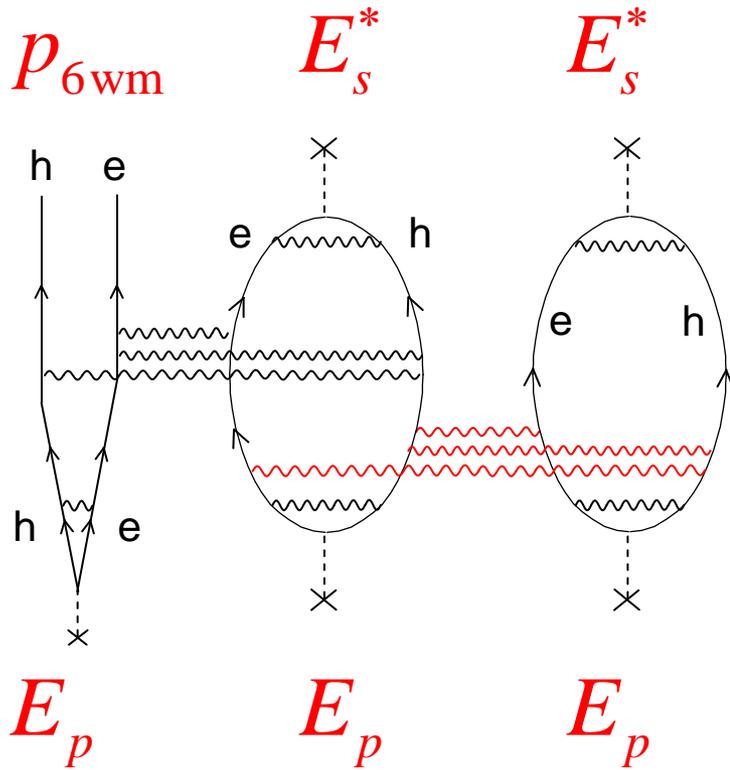
"incoh. dens.  
assist. trans."

plus:

- renormalization of  $V$  or  $G$
- triexciton

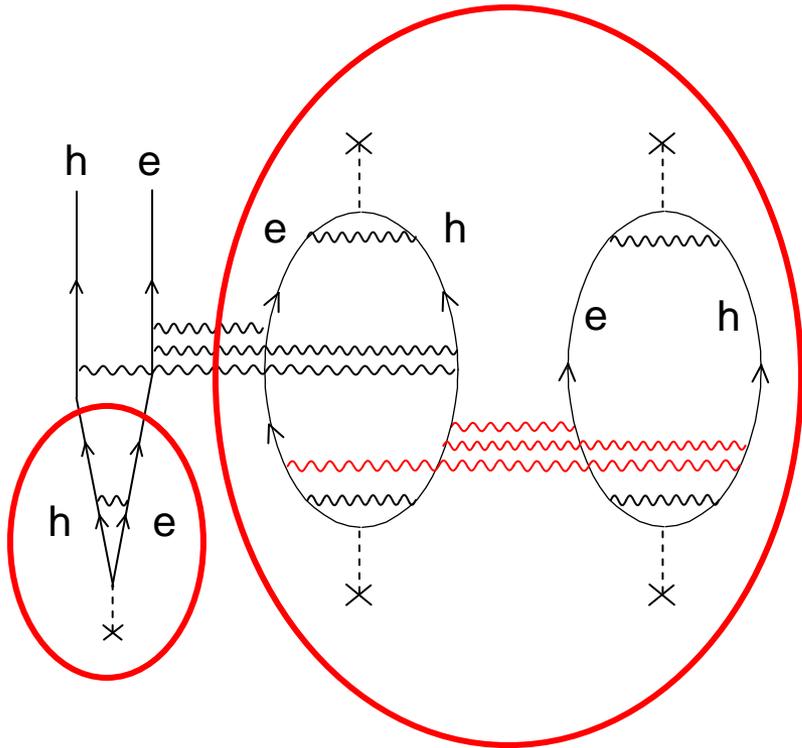


## Six Wave Mixing

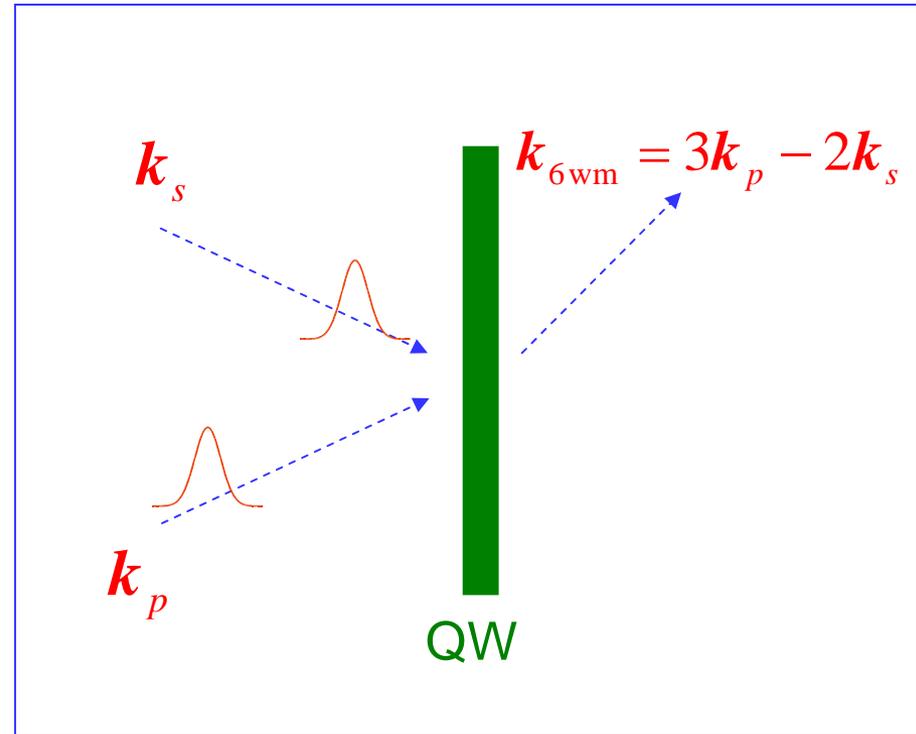




## Six Wave Mixing



$[Gnp]$





## Demonstration of Sixth-Order Coulomb Correlations in a Semiconductor Single Quantum Well

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(Received 9 March 2000)

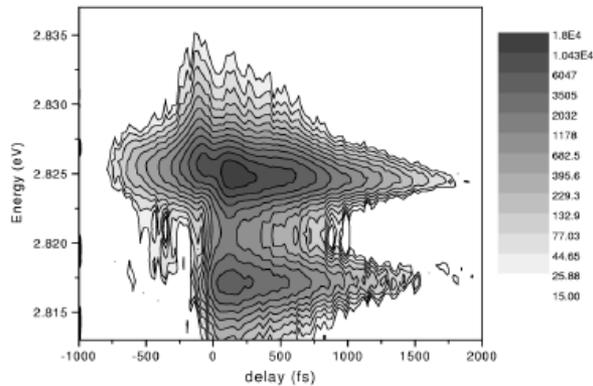


FIG. 1. Six-wave mixing emission measured in the  $\{X, Y, Y\}$  configuration, shown over 3 orders of magnitude in emission intensity.

Experiment

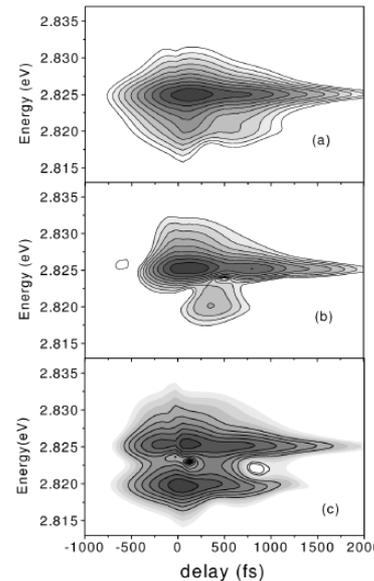


FIG. 2. Theoretical calculations of SWM emission in the  $\{X, Y, Y\}$  configuration, shown over 3 orders of magnitude in emission intensity: (a) calculation in the coherent limit CL including  $Y$  and  $\bar{B}$ ; (b) calculation including contributions of  $Y$ ,  $\bar{B}$ , and  $N$ , i.e., the ID level; (c) calculation on the IDAT level including the four- and six-point contributions  $Y$ ,  $\bar{B}$ ,  $\bar{N}$ , and  $Z$ .

Theory

"coherent limit"

"incoherent densities"

"incoherent-density assisted transitions"

## Biexcitonic effects in the coherent control of the excitonic polarization detected in six-wave-mixing signals

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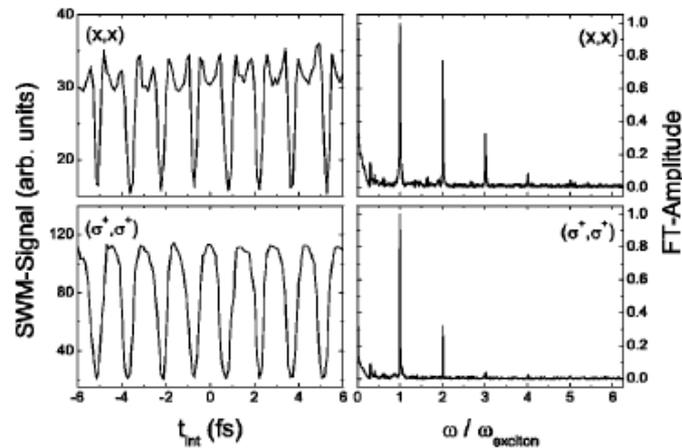


FIG. 2. Interferometric SWM signal measured for  $(x,x)$  colinearly (top, with biexcitonic contributions) and  $(\sigma^+, \sigma^+)$  cocircularly (bottom, without biexcitonic contributions) polarized incident beams at  $t_{del} = 0.6$  ps. The corresponding FT spectra (right) are each normalized to the value of  $\omega_{exc}$ .

Experiment

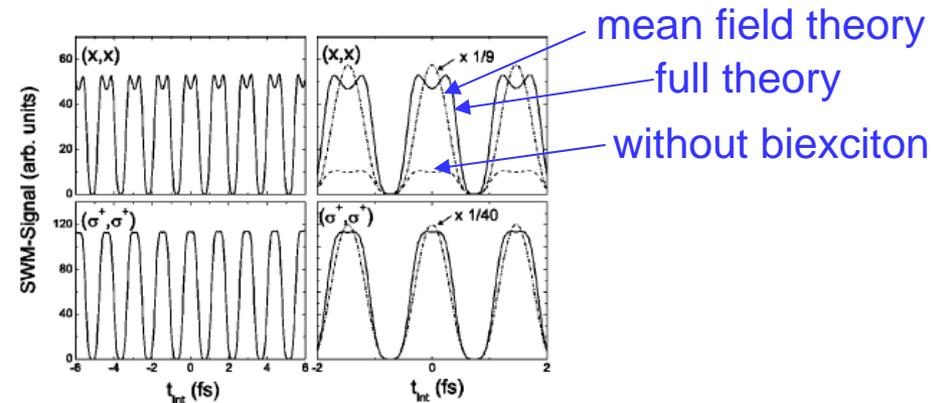


FIG. 3. Calculated interferometric SWM signal for  $(x,x)$  colinearly (top) and  $(\sigma^+, \sigma^+)$  cocircularly (bottom) polarized incident beams. Left panels: full theory; right panels: (solid) full theory, (dashed) full theory without biexciton contributions, and (dot-dashed) mean-field theory scaled by the indicated factors. Parameters used in the calculations: electron mass  $m_e = 0.14m_0$ , hole mass  $m_h = 0.67m_0$ ,  $m_0$  = free-electron mass, static dielectric constant  $\epsilon_s = 8.7$ , interband dephasing time  $T_2 = 3.3$  ps, and pulse area  $A = 0.07$  for all three pulses.

Theory



## Light-polarization and intensity dependence of higher-order nonlinearities in excitonic FWM signals

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<sup>1</sup> Institute for Theoretical Physics, University of Bremen, 28334 Bremen, Germany

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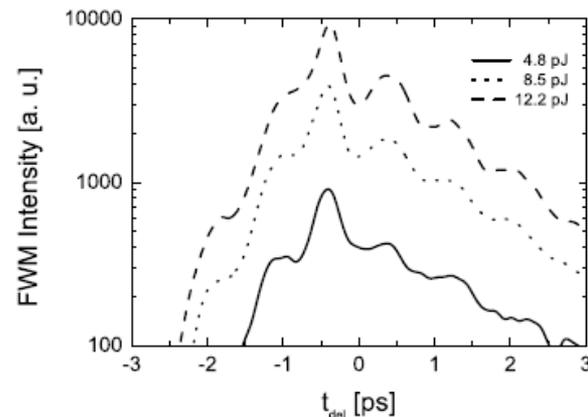


Fig. 2. Experiment: FWM signal vs. delay time for equal intensities of both pulses which are linearly polarized enclosing an angle of  $\phi_{\text{pol}} = 75^\circ$ . The pulse energies are varied from 4.8 to 12.2 pJ. Signal detection is at the spectral position of the excitonic resonance.

Experiment

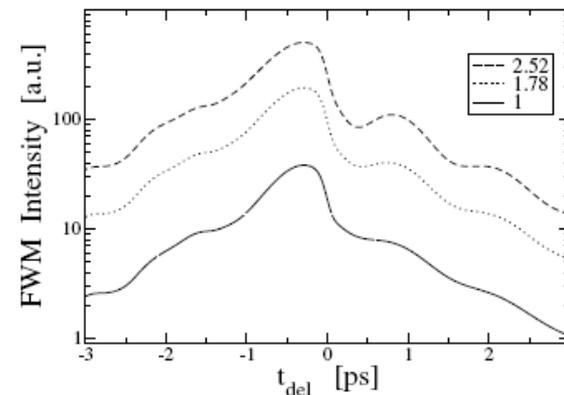


Fig. 5. Theoretical result for the same excitation and detection conditions as in Figure 2. The lowest intensity corresponds to a Rabi energy of  $dE/E_B = 6.3 \times 10^{-3}$  in units of the  $3d$  excitonic Rydberg energy  $E_B$ .

Theory



## Spectral signatures of $\chi^{(5)}$ processes in four-wave mixing of homogeneously broadened excitons

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*Research Center COM, Technical University of Denmark, Building 349, 2800 Lyngby, Denmark*

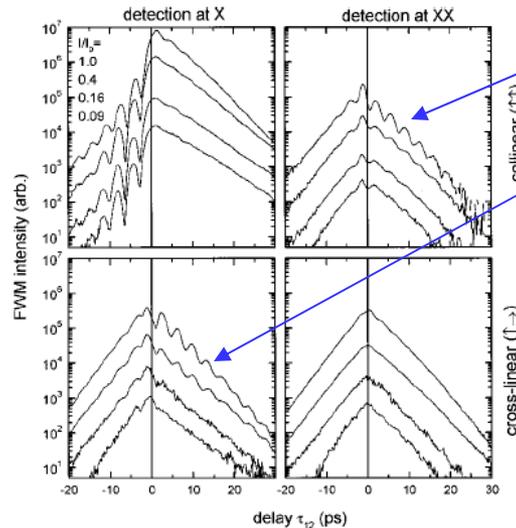


Fig. 4. FWM traces for various excitation intensities, as labeled ( $I_0 = 300 \text{ nJ/cm}^2$ ). All traces are scaled by the same factor. Top, collinearly polarized excitation ( $\uparrow\uparrow$ ); bottom, cross-linearly polarized excitation ( $\uparrow\rightarrow$ ); left, detection energy at the exciton resonance (X); right, detection energy at the exciton-biexciton resonance (X-XX).

Experiment

beats with inverse biexciton binding energy period

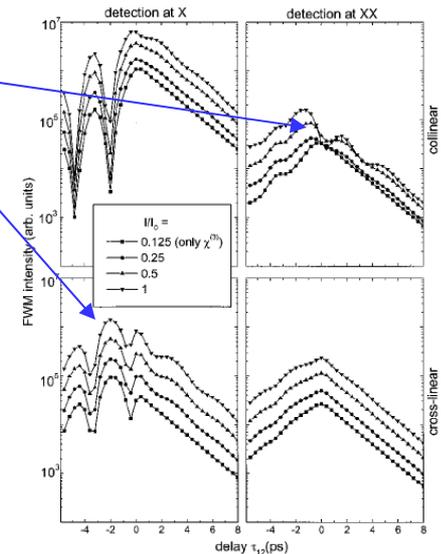


Fig. 6. Calculated FWM traces for various excitation intensities, as labeled. Top, collinearly polarized excitation ( $\uparrow\uparrow$ ); bottom, cross-linearly polarized excitation ( $\uparrow\rightarrow$ ); left, detection energy at the exciton resonance (X); right, detection energy at the exciton-biexciton resonance (X-XX). The symbols indicate the calculated time delays; the lines are guides for the eye.

Theory



# Spectral signatures of $\chi^{(5)}$ processes in four-wave mixing of homogeneously broadened excitons

W. Langbein

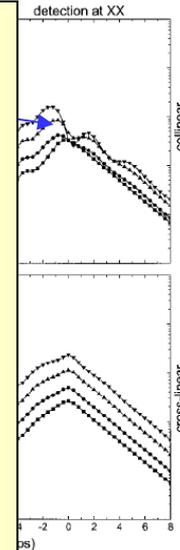
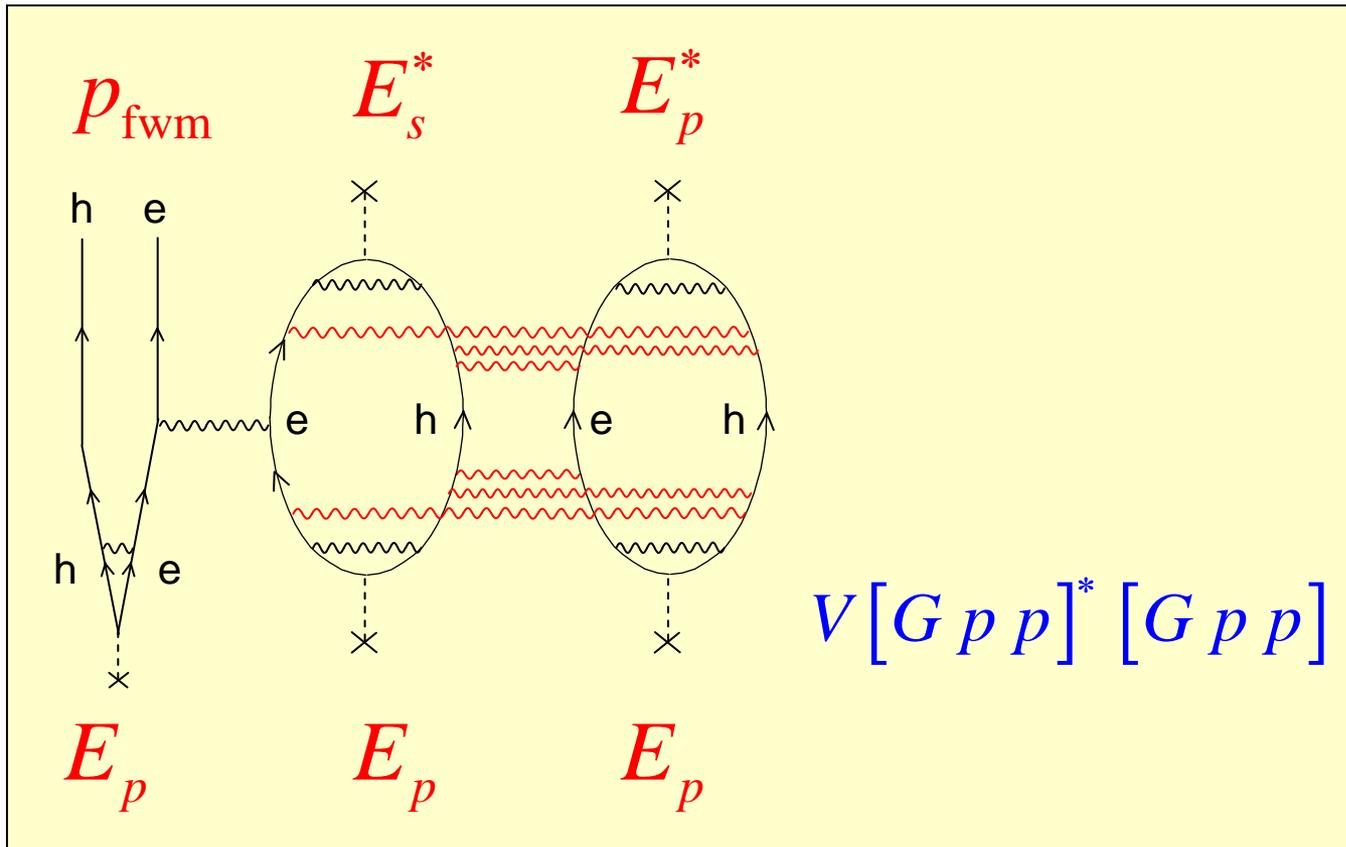
*Experimentelle Physik E11b, Universität Dortmund, Otto-Hahn Strasse 4, 44227 Dortmund, Germany*

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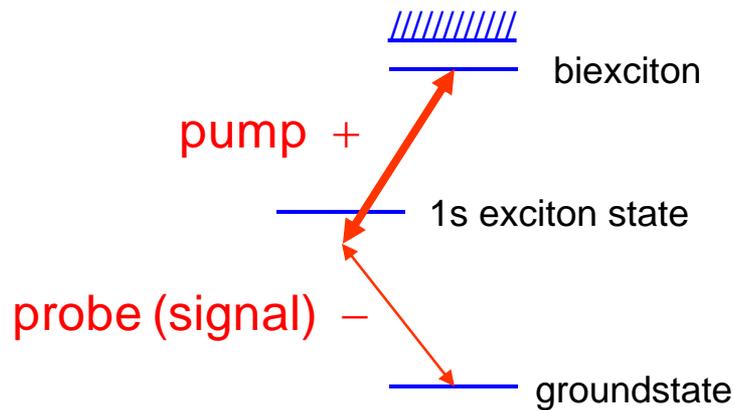


various excitation intensities (↑↓); bottom (↑→); left, detection energy at the... The symbols indicate the guides for the eye.

ory



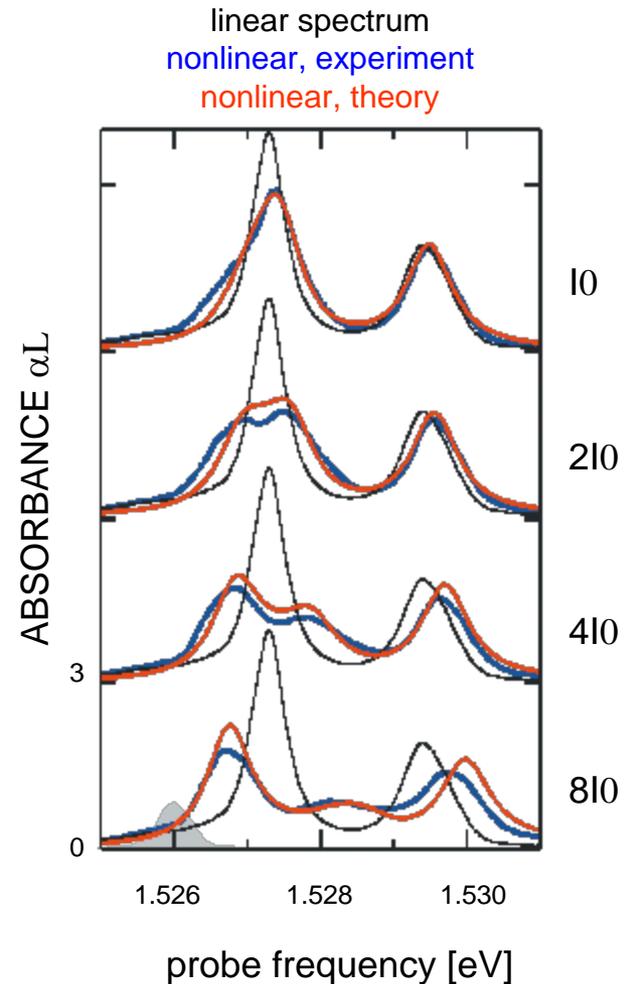
## Electromagnetically-induced transparency (EIT)



EIT dip at

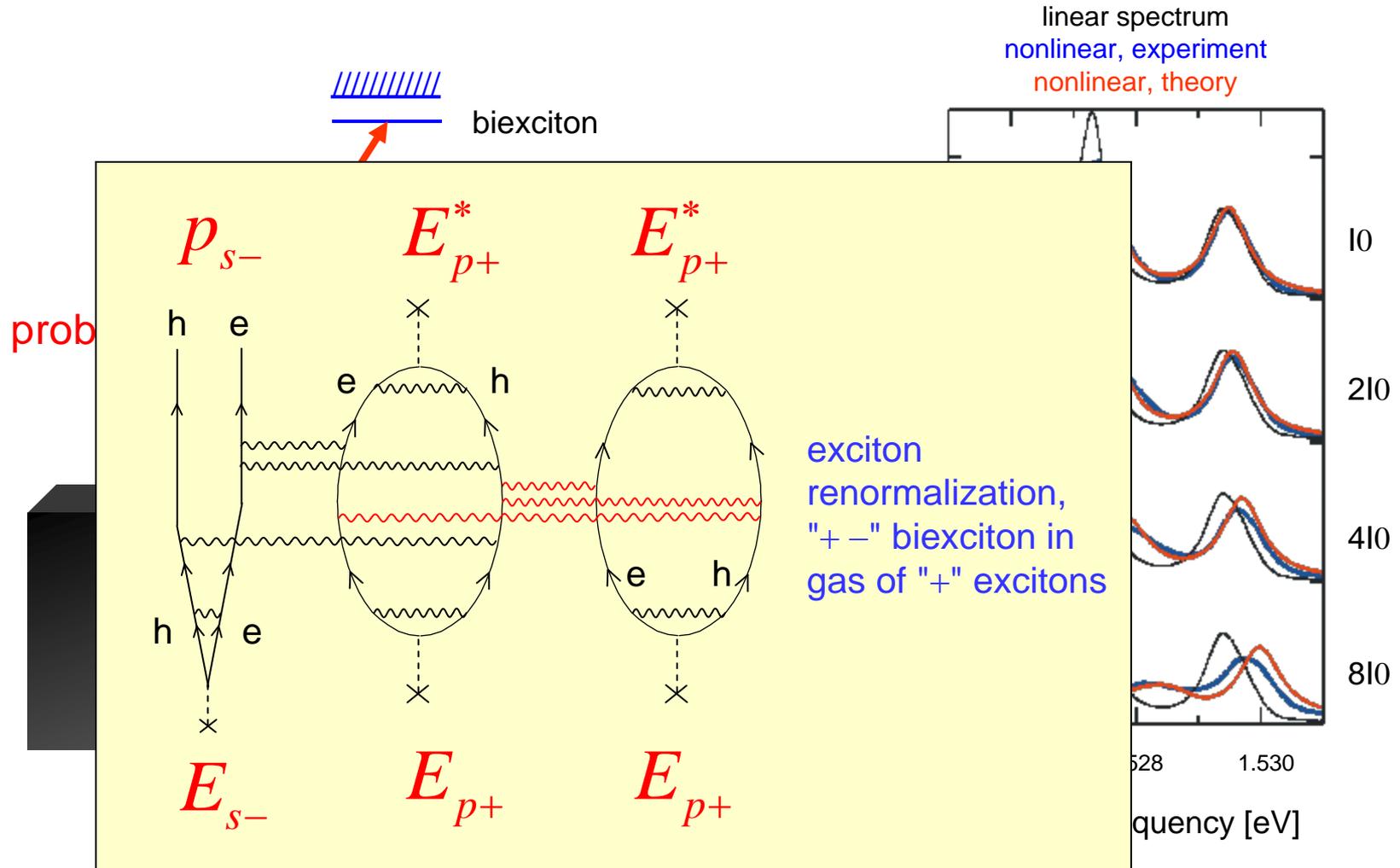
$$\hbar\omega_{pump} + \hbar\omega_{probe} \approx \varepsilon_{biexciton}$$

shifts with increasing pump intensity



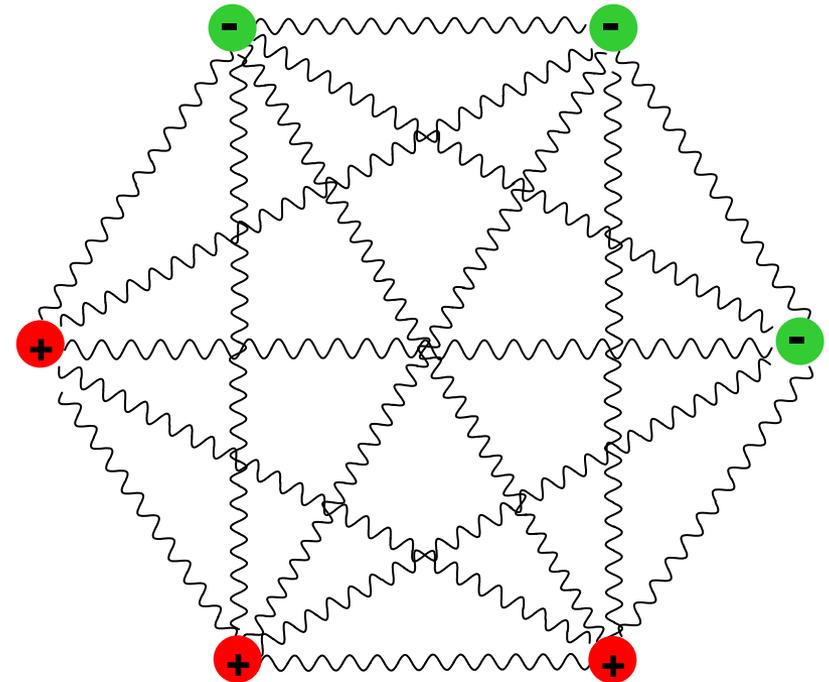
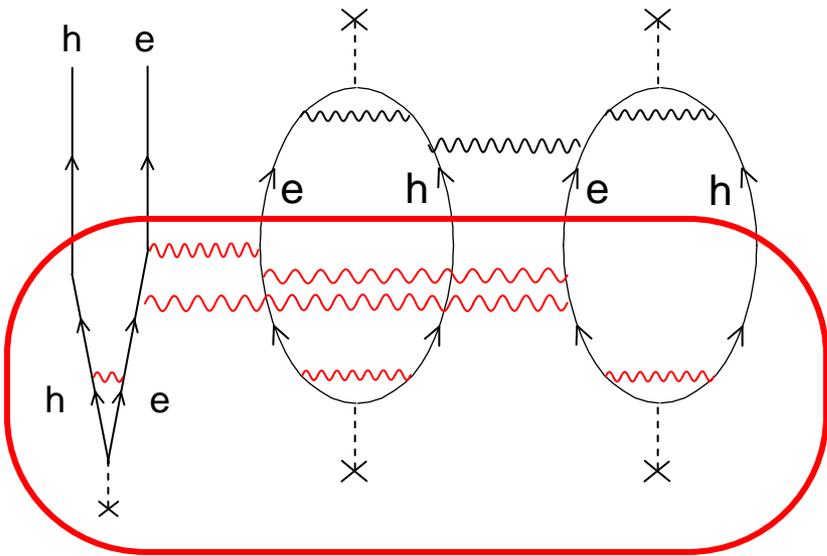


# Electromagnetically-induced transparency (EIT)





## Triexciton states?



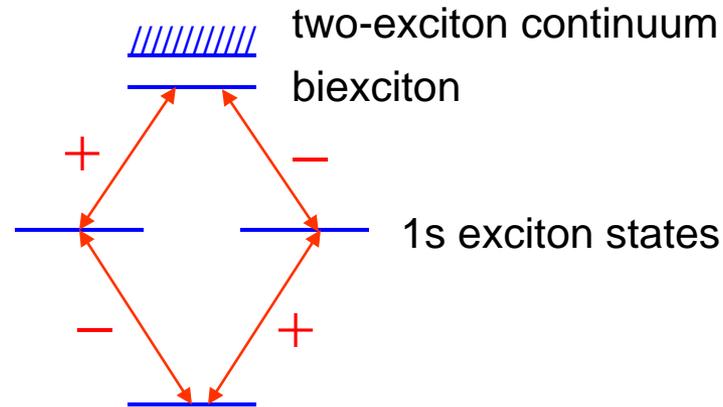
$$i \frac{d}{dt} \langle a a \rangle \Big|_{corr}$$
$$= V \langle a^\dagger a^\dagger a^\dagger a^\dagger \rangle \langle a a a a a a \rangle_{corr}$$

triexciton



*Next:*

Few-level systems



## Few level models:

- are useful for conceptual analysis of optical nonlinearities
- can be used in "double-sided Feynman diagrams"  
(e.g. in Li, Zhang, Borca, Cundiff, Phys. Rev. Lett. 96, 057406 (2006))

Question: can they be strictly related to many-particle theory?

Answer: yes, at least in  $\chi^{(3)}$  regime!



## Many-Body Interactions in Semiconductors Probed by Optical Two-Dimensional Fourier Transform Spectroscopy

Xiaoqin Li,<sup>1</sup> Tianhao Zhang,<sup>1,2</sup> Camelia N. Borca,<sup>1</sup> and Steven T. Cundiff<sup>1</sup>

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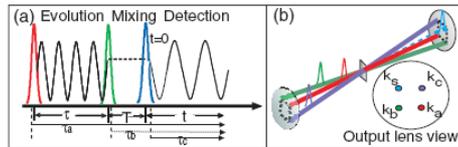


FIG. 1 (color). (a) Schematic of the excitation sequence showing the relevant time intervals. Note the dashed line indicating that the phase during time period  $t$  is related to the phase evolution during time  $\tau$ . (b) Diagram showing geometry of excitation pulses and generated signal.

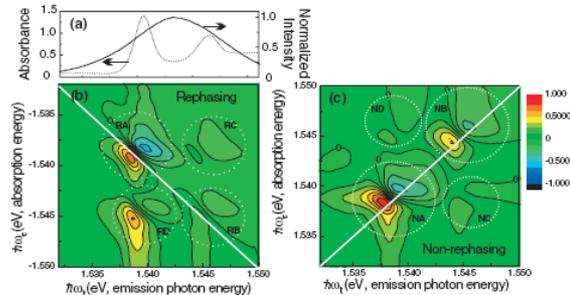


FIG. 3 (color). Linear absorption, excitation pulse spectrum (a) and experimental real spectra for the rephasing (b) and nonrephasing (c) pulse sequences. Both spectra in (b) and (c) are normalized to the most intense peak.

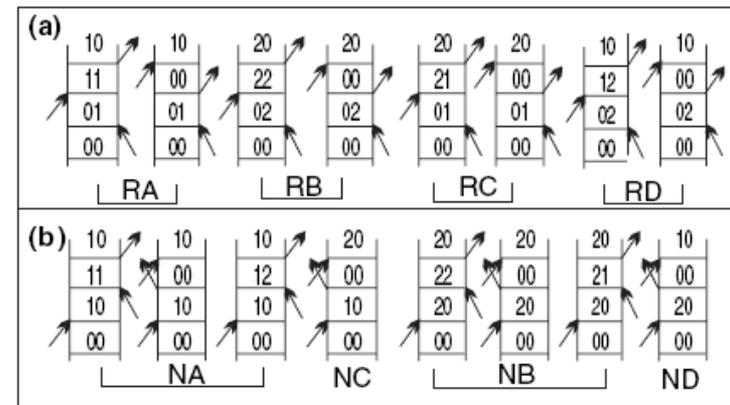
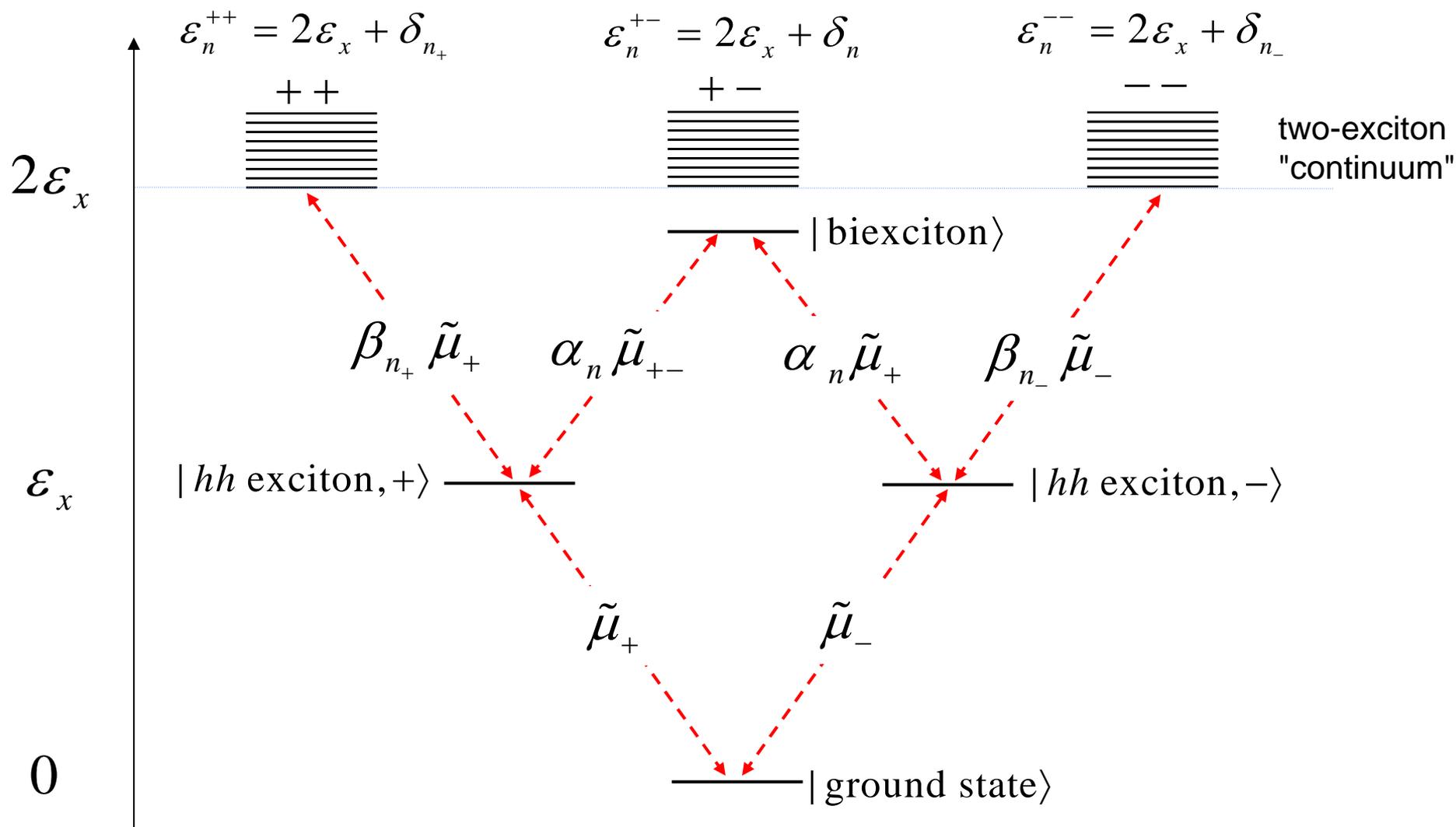


FIG. 2. Feynman diagrams for all possible quantum mechanical pathways for (a) rephasing and (b) nonrephasing measurements in a V system, where 0, 1, 2 corresponds to crystal ground state, heavy-hole, and light-hole excitons, respectively.

see Robert W. Boyd, *Nonlinear Optics*,  
(Academic Press, London, 1992)





$$i\hbar \frac{d}{dt} \rho_{ij} = \sum_k \left( H_{ik} \rho_{kj} - \rho_{ik} H_{kj} \right)$$

Initial condition:  
system in ground state

$$P_+(t) = N_a \tilde{\mu}_+^* p_+(t)$$

↑  
density of few-level 'atoms'

$$p_+(t) = \rho_{\text{exc}+,g}(t) + \sum \alpha_n^* \rho_{n,\text{exc}+}(t) + \sum \beta_n^* \rho_{n_+,\text{exc}+}(t)$$



$$\begin{aligned}
 i\hbar \dot{p}^+ &= (\varepsilon_x - i\gamma) p^+ - [1 - 2\nu |p^+|^2] \tilde{E}^+ \\
 &\quad + V_{+-}^{phen} |p^-|^2 p^+ \\
 &\quad + V_{++}^{phen} |p^+|^2 p^+ \\
 &\quad + 2 p^{+*} \int_{-\infty}^{\infty} dt' G_{++}^{phen}(t-t') p^+(t') p^+(t') \\
 &\quad + p^{-*} \int_{-\infty}^{\infty} dt' G_{+-}^{phen}(t-t') p^+(t') p^-(t') \\
 &\quad + \text{terms proportional to } \gamma\text{'s}
 \end{aligned}$$

$$i\hbar \dot{p}^+ = (\varepsilon_x - i\gamma) p^+ - [\phi_{1s}^*(0) - 2A^{\text{PSF}} |p^+|^2] E^+$$

Phase-space filling

$$+ V^{\text{HF}} |p^+|^2 p^+$$

Hartree-Fock  
Coulomb interaction

$$+ 2 p^{+*} \int_{-\infty}^{\infty} dt' G^{++}(t-t') p^+(t') p^+(t')$$

$$+ p^{-*} \int_{-\infty}^{\infty} dt' G^{+-}(t-t') p^+(t') p^-(t')$$

} Time-retarded  
two-exciton  
correlations  
(incl. biexciton)



We have set  $\sum |\alpha_n|^2 = 1$

$$\nu = 1 - \frac{1}{2} \sum |\beta_n|^2$$

$$V_{++}^{phen} = \frac{1}{2} \sum |\beta_n|^2 \delta_{n_+}$$

$$V_{+-}^{phen} = \sum |\alpha_n|^2 \delta_n$$

$$G_{++}^{phen}(t-t') = \left(-\frac{i}{\hbar}\right) \theta(t-t') \sum \frac{\beta_n^* \delta_{n_+}}{2} e^{-\frac{i}{\hbar}(2\varepsilon_x + \delta_{n_+} - i\gamma_b)(t-t')} \frac{\beta_n \delta_{n_+}}{2}$$

$$G_{+-}^{phen}(t-t') = \left(-\frac{i}{\hbar}\right) \theta(t-t') \sum \alpha_n^* \delta_n e^{-\frac{i}{\hbar}(2\varepsilon_x + \delta_n - i\gamma_b)(t-t')} \alpha_n \delta_n$$



## Identification of few-level parameters

$$N_a |\tilde{\mu}|^2 \leftrightarrow |\phi_{1s}(0)|^2 |\mu|^2$$

$$\frac{1}{N_a} v \leftrightarrow A^{PSF} / |\phi_{1s}(0)|$$

$$\frac{1}{N_a} V_{++}^{phen} \leftrightarrow V^{HF}$$

$$\frac{1}{N_a} V_{+-}^{phen} \leftrightarrow 0$$

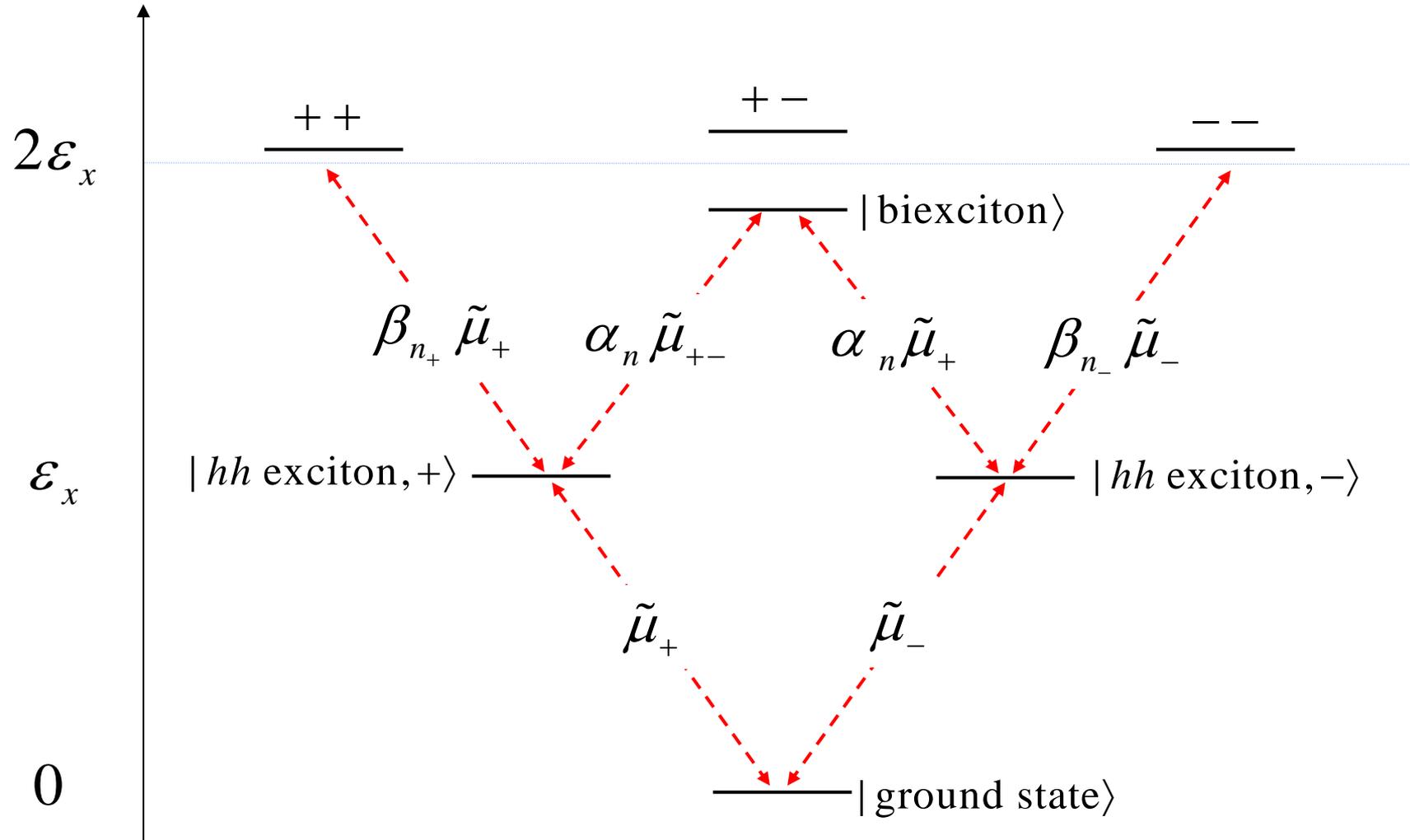
$$\left. \frac{1}{N_a} G_{++}^{phen}(t-t') \leftrightarrow G_{++}(t-t') \right\}$$

$$\left. \frac{1}{N_a} G_{+-}^{phen}(t-t') \leftrightarrow G_{+-}(t-t') \right\}$$

← approximate  
(more 'atomic' levels  
yield better agreement)

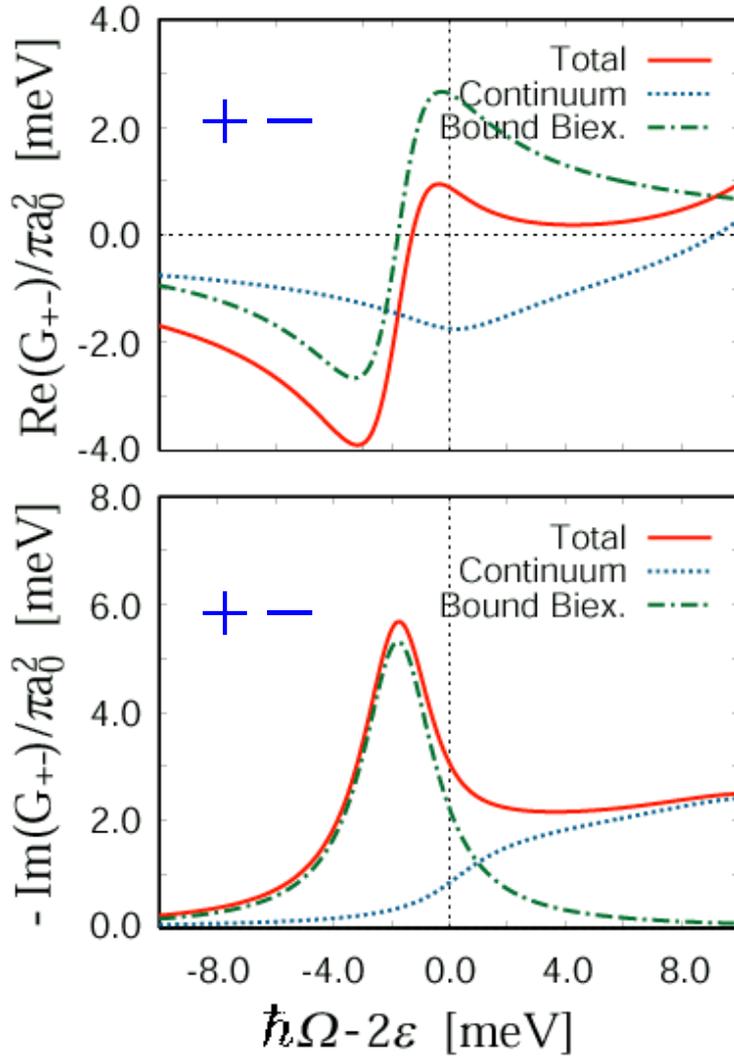


## Example: 7-level system

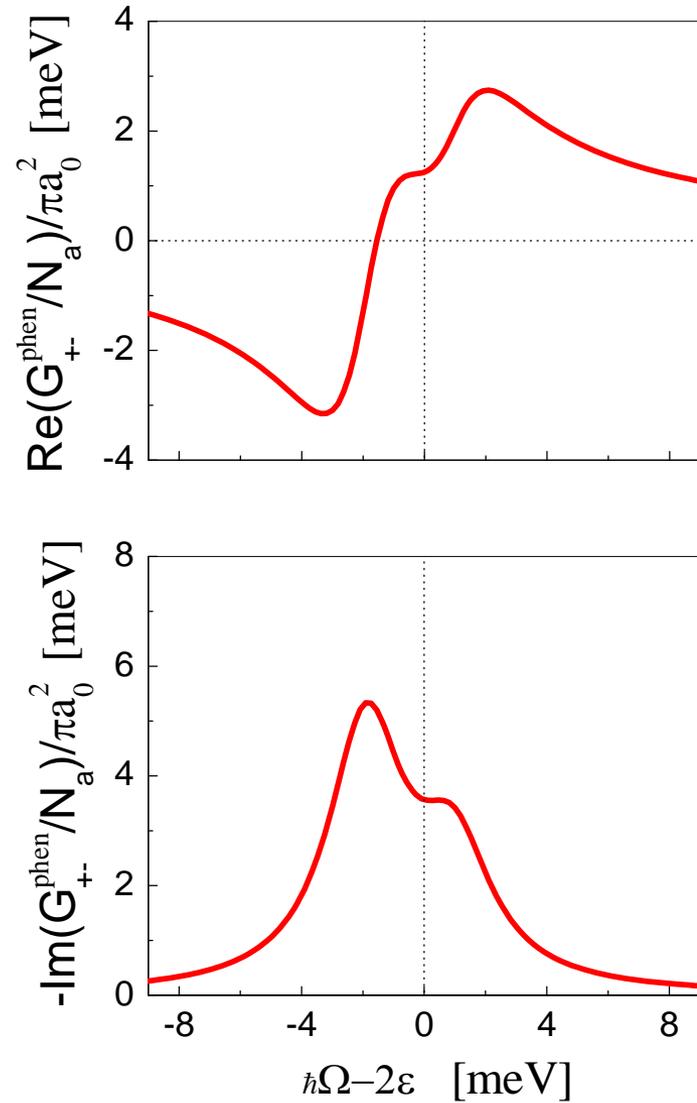




## Microscopic theory

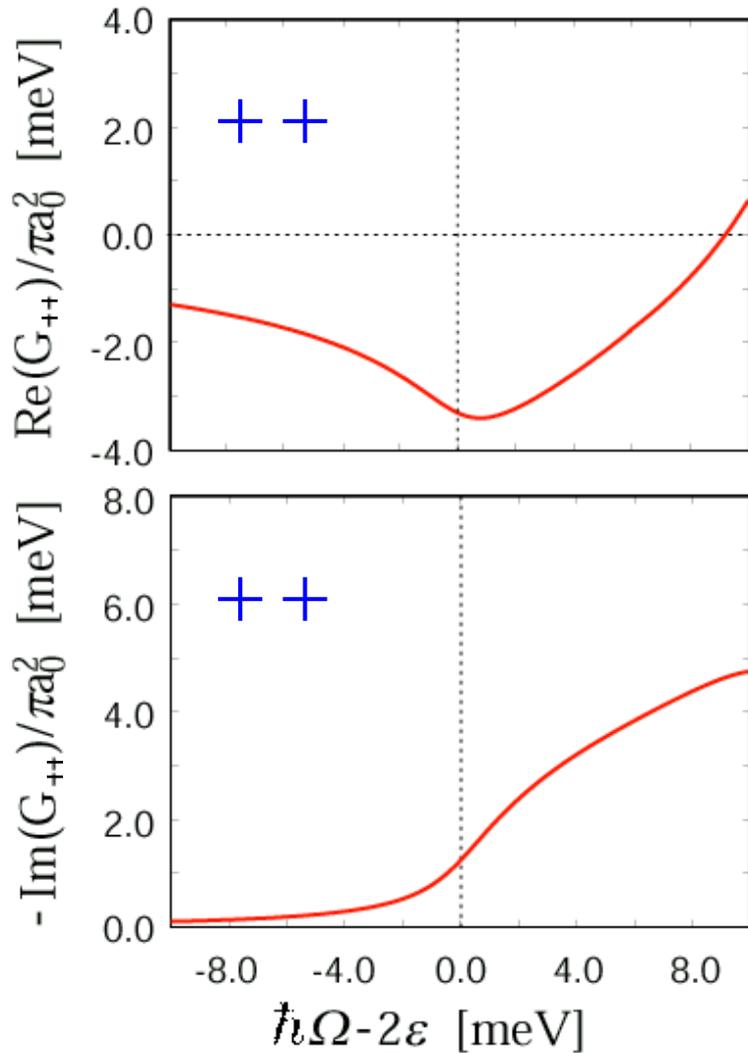


## 7-level system

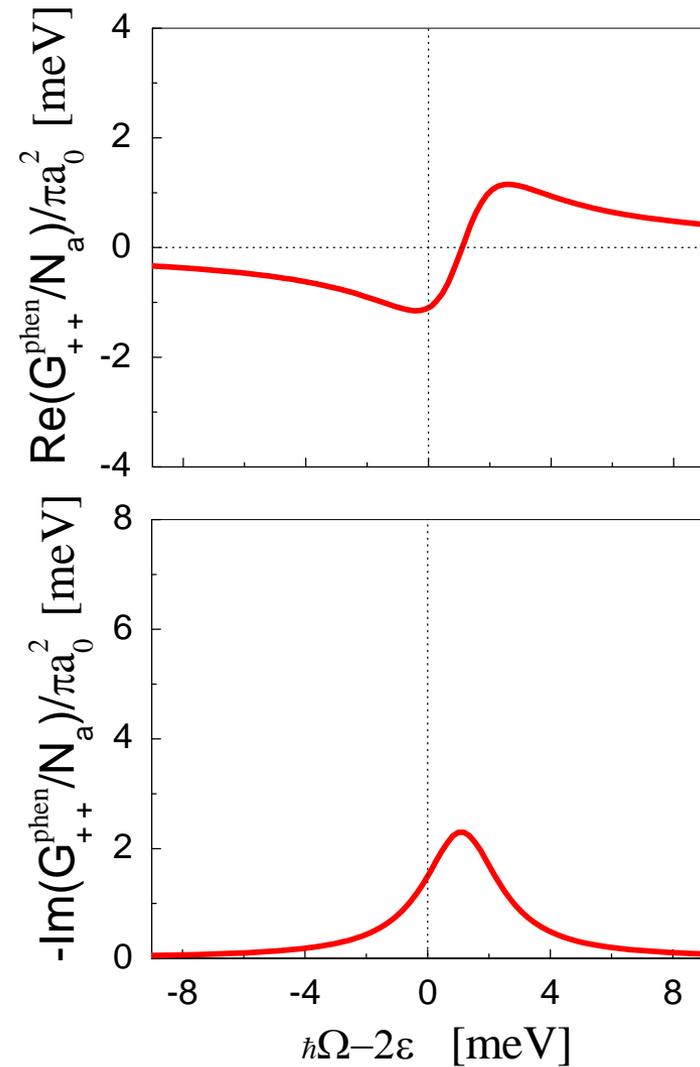




## Microscopic theory



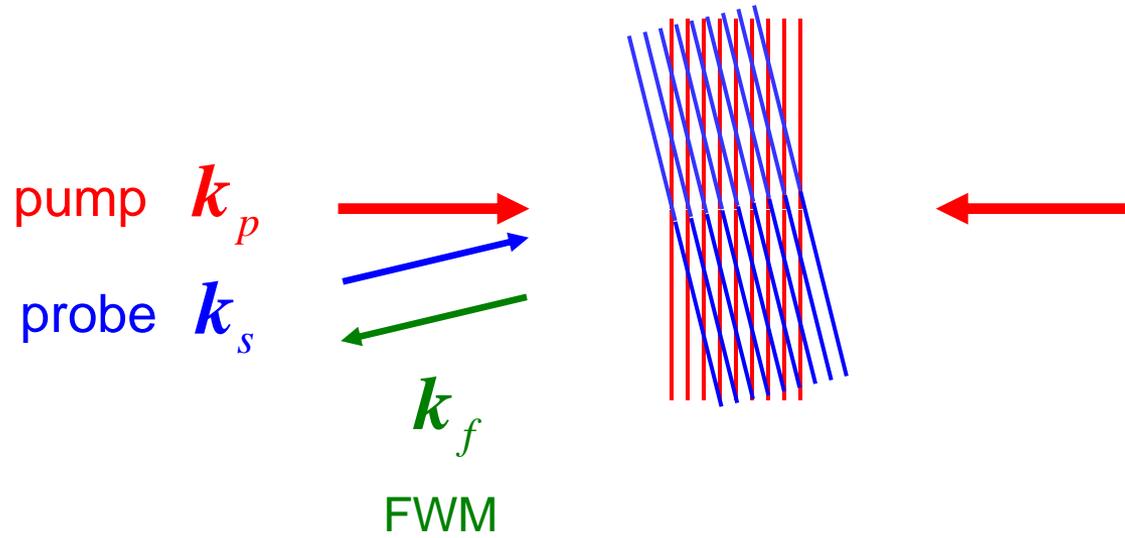
## 7-level system

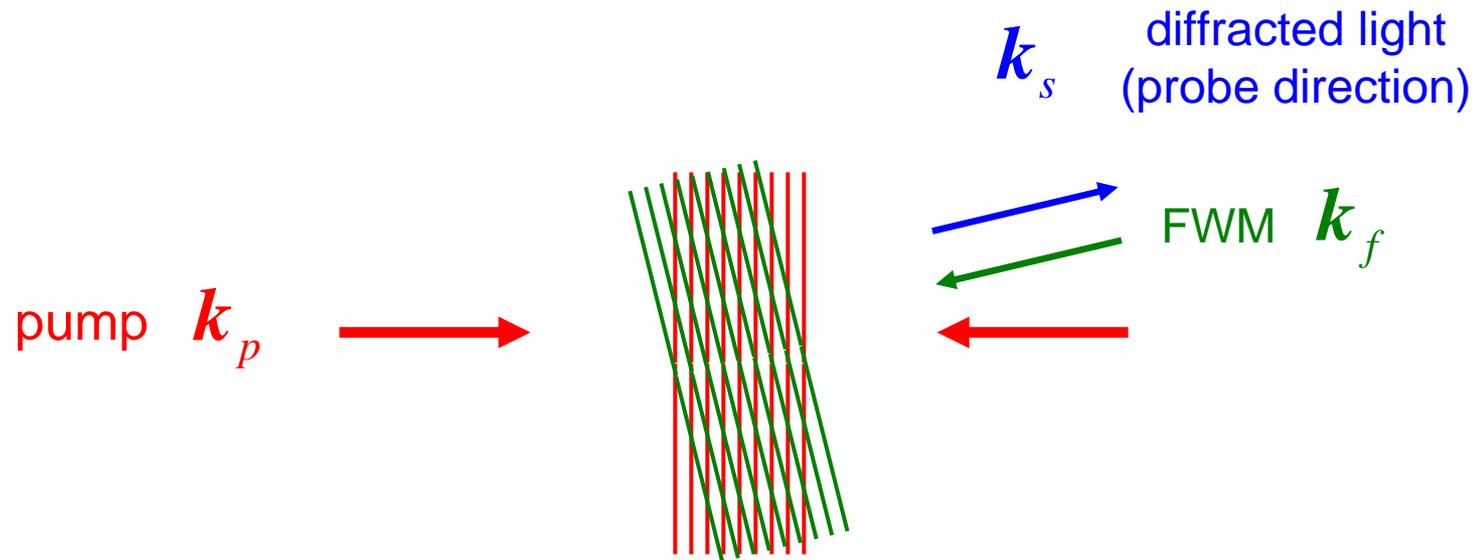


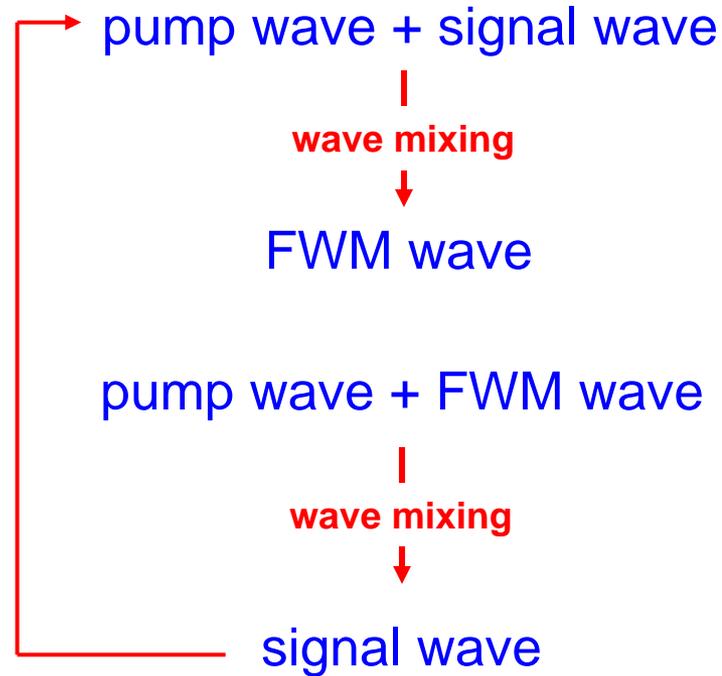


*Next:*

FWM instabilities







**feedback and possible dynamic instability**



$$\begin{aligned} i\hbar \dot{p}_f &= V^{HF} p_p p_p p_s^* \\ i\hbar \dot{p}_s &= V^{HF} p_p p_p p_f^* \end{aligned}$$

self consistency (positive feedback)

assume  $p_p(t) = \tilde{p}_p e^{-i\omega_p t}$  and  $p_s(t) = \tilde{p}_s(t) e^{-i\omega_p t}$   
 $p_f(t) = \tilde{p}_f(t) e^{-i\omega_p t}$

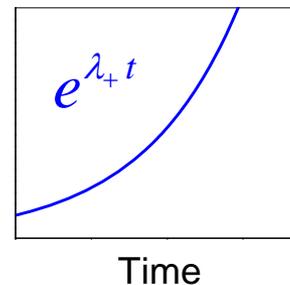
$$\dot{\mathbf{A}} = \mathbf{M} \mathbf{A}$$

with  $\mathbf{A} = \begin{pmatrix} \tilde{p}_s \\ \tilde{p}_f^* \end{pmatrix}$   $\mathbf{M} = \begin{pmatrix} 0 & -i\kappa \\ i\kappa & 0 \end{pmatrix}$   $\kappa = V^{HF} \tilde{p}_p^2$

### Linear stability analysis:

$$\mathbf{A} = \mathbf{A}_0 e^{\lambda t}$$

$$\det(\mathbf{M} - \lambda) = 0$$



$$\lambda_{\pm} = \pm |\kappa|$$



## Stimulated polariton scattering in semiconductor microcavities

- Savvidis, Baumberg, Stevenson, Skolnick, Whittaker, and Roberts, Phys. Rev. Lett. 84, 1547 (2000)
- Huang, Tassone, Yamamoto, Phys. Rev. B 61, R7854 (2000)
- Ciuti, Schwendimann, Deveaud, Quattropani, Phys. Rev. B 62, R4825 (2000)
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- Kasprzak, Richard, Kundermann, Baas, Jeambrun, Keeling, Marchetti, Szymanska, Andre, Staehli, Savona, Littlewood, Deveaud, LeSiDang, Nature 443, 409 (2006)

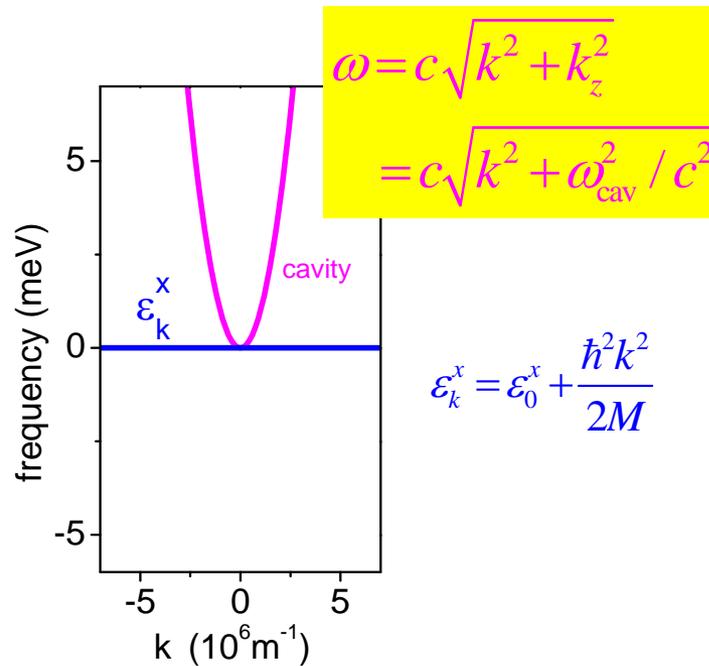
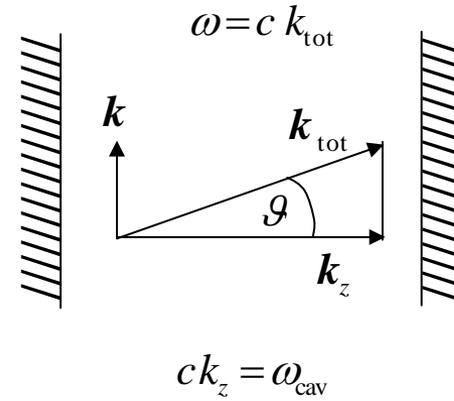
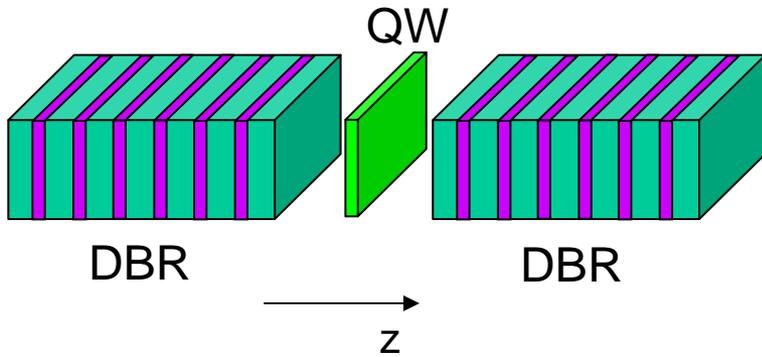
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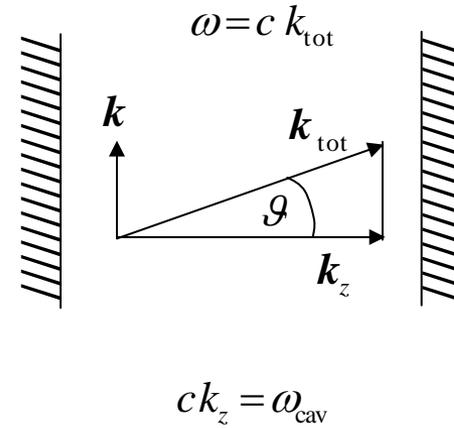
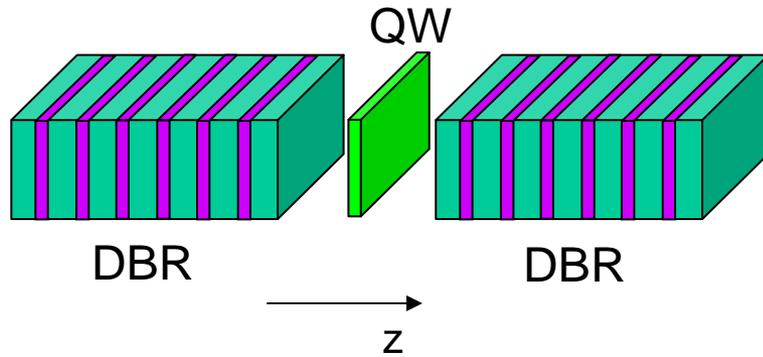


## Stimulated polariton scattering in semiconductor microcavities

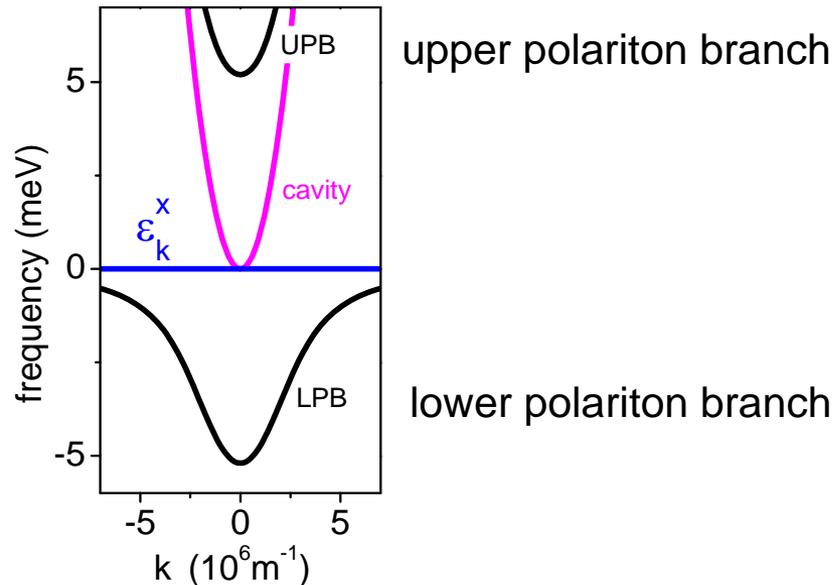
- Savvidis, Baumberg, Stevenson, Skolnick, Whittaker, and Roberts, Phys. Rev. Lett. 84, 1547 (2000)
  - Huang, Tassone, Yamamoto, Phys. Rev. B 61, R7854 (2000)
  - Ciuti, Schwendimann, Deveaud, Quattropani, Phys. Rev. B 62, R4825 (2000)
  - Stevenson, Astratov, Skolnik, Whittaker, Tassone, Ciuti, Baumberg, Roberts, Phys. Rev. B 62, R4825 (2000)
  - Savasta, DiStefano, Girlanda, Phys. Rev. B 62, R4825 (2000)
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  - Klotz, Klotz, Martin, Amo, Vina, Shlykh, Glazo, Malpuech, Kavokin, Andre, Solid State Commun. 139, 511 (2006)
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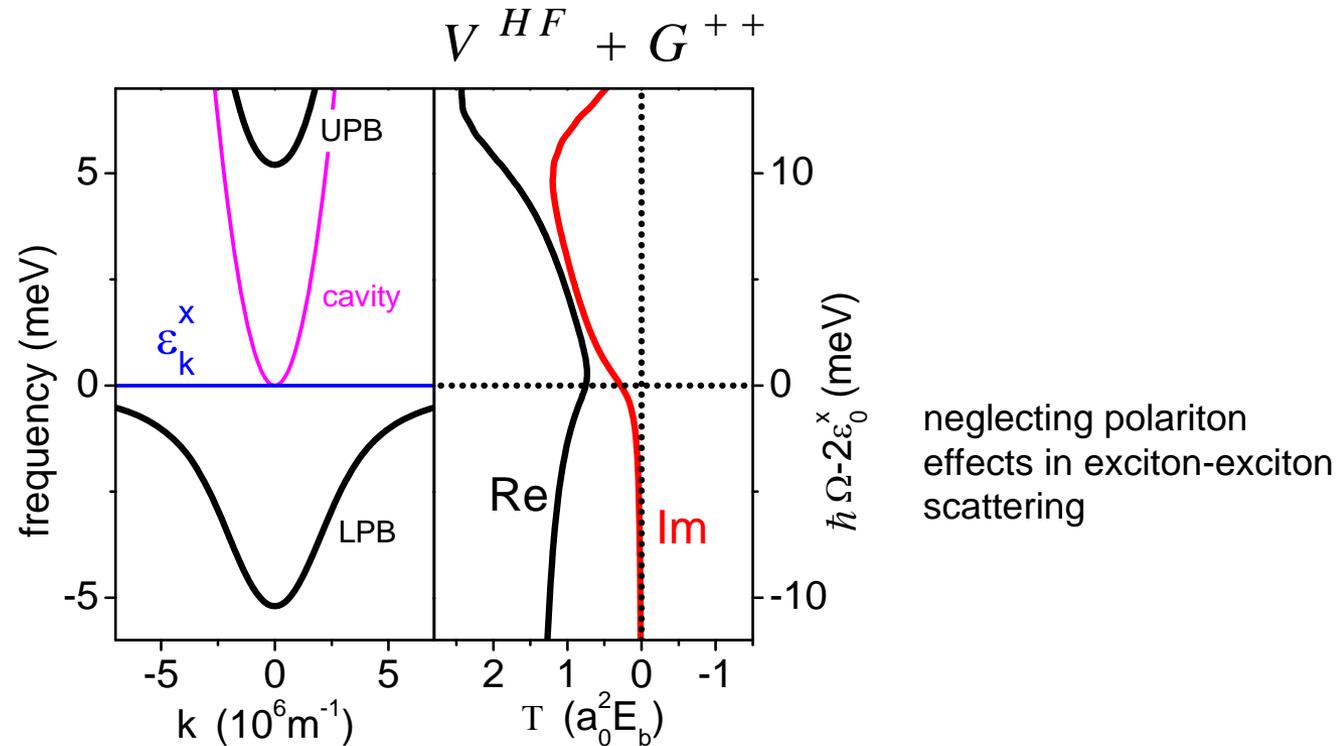
The publications in 2000 spurred major activities in bosonic aspects of excitons. In this tutorial, only FWM aspects are covered, not the bosonic aspects.





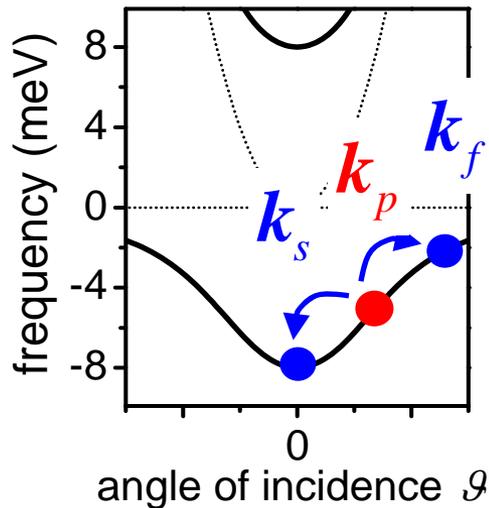
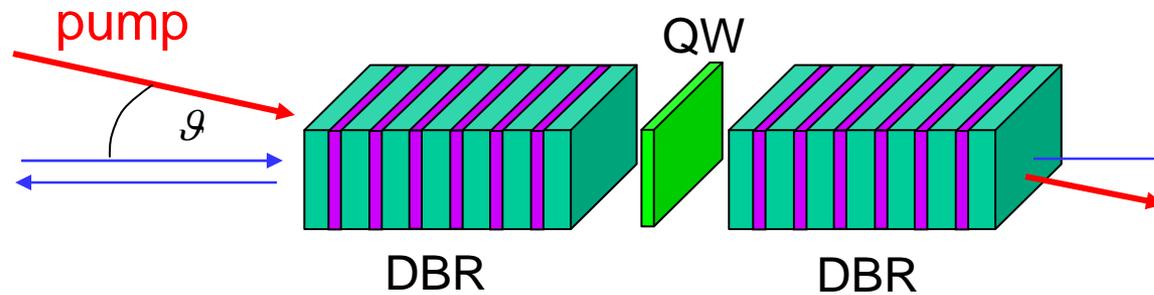
Coupled modes:





- On LPB, two-exciton correlation dominated by Hartree-Fock
- Small excitation-induced dephasing at LPB facilitates instability (in contrast to single quantum well<sup>(\*)</sup>)

- Savasta, DiStefano, Girlanda, Phys. Rev. Lett. 90, 096403 (2003)
- Schumacher, Kwong, Binder, Phys. Rev. B 76, 245324 (2007)
- (\*)Schumacher, Kwong, Binder, Europhys. Lett. 81, 27003 (2008)
- (\*)Schumacher, Kwong, Binder, Smirl, Phys. Stat. Sol. (b) 246, 307 (2009)

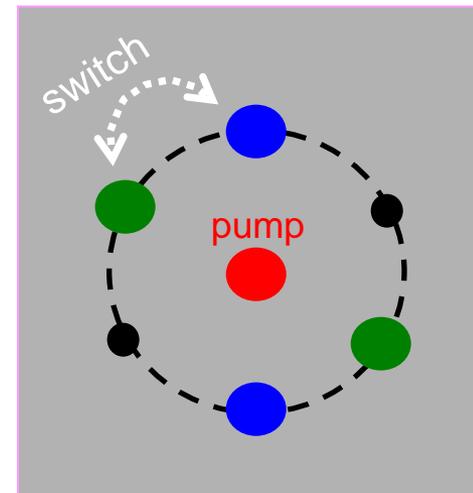
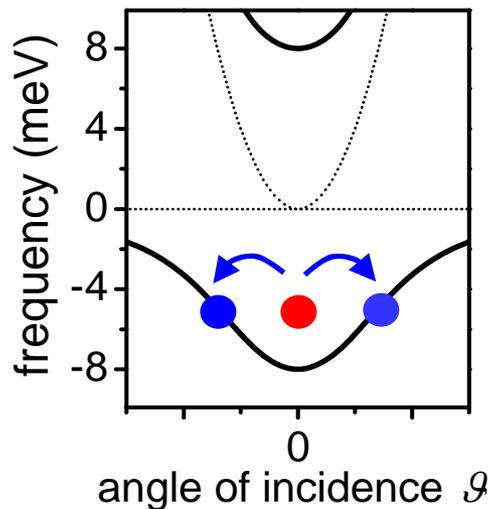
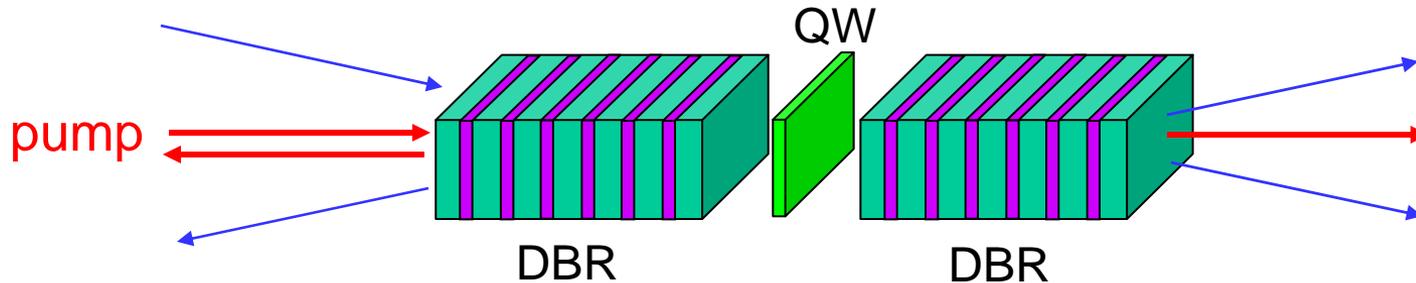


instability threshold low at "magic angle"

- Savvidis, Baumberg, Stevenson, Skolnick, Whittaker, and Roberts, Phys. Rev. Lett. 84, 1547 (2000)
- Huang, Tassone, Yamamoto, Phys. Rev. B 61, R7854 (2000)
- Ciuti, Schwendimann, Deveaud, Quattropani, Phys. Rev. B 62, R4825 (2000)



## Low-intensity directional manipulation semiconductor proposal for low-intensity switch demonstrated in Dawes, Illing, Clark, Gauthier, Science 308, 672 (2005)



- Schumacher, Kwong, Binder, Smirl, Phys. Stat. Sol. RRL 3, 10 (2009)
- Dawes, Gauthier, Schumacher, Kwong, Binder, Smirl, Laser & Photon. Rev. (2009)



*Last:*

Conclusion



During the last 20 years, FWM techniques developed by D.S. Chemla and others have given us deep insight into many-particle processes in optically excited semiconductors, including the observation of excitonic correlation effects.

The physical processes underlying these effects can be visualized (and analyzed) with the help of Feynman diagrams.

This tutorial talk is available at  
*[www.optics.arizona.edu/binder](http://www.optics.arizona.edu/binder)*