Seminar on optical properties of semiconductors

WAVE MIXING SPECTROSCOPY

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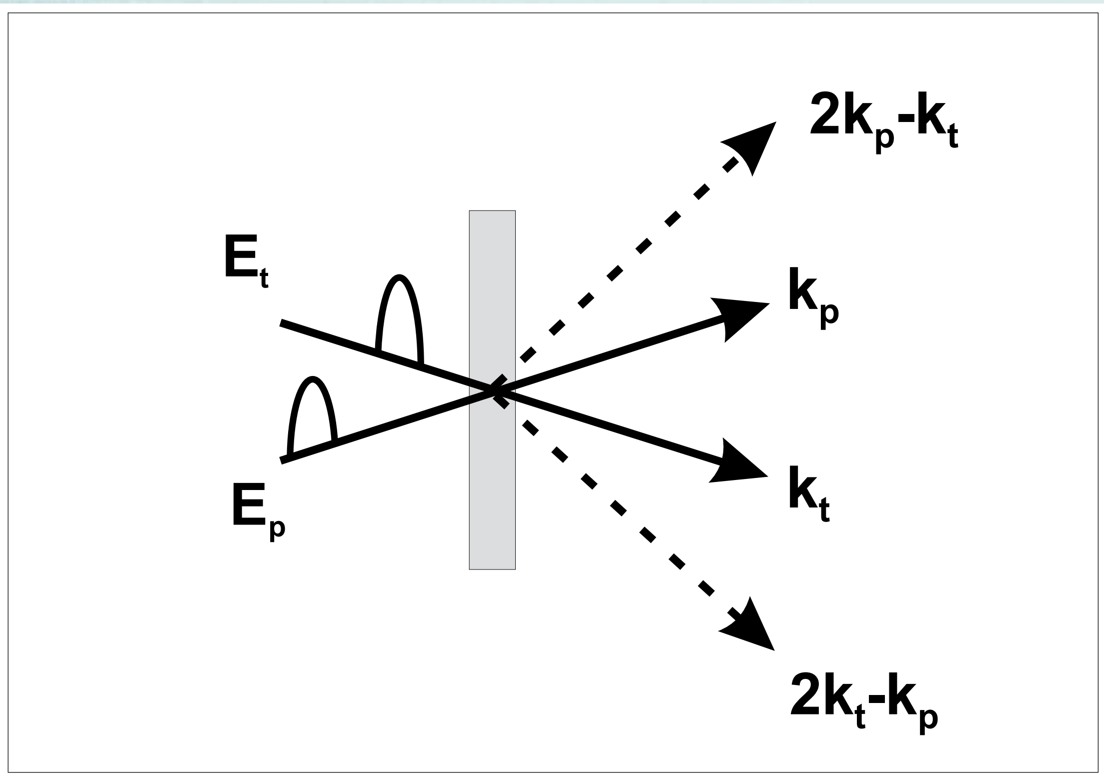
TWO-PULSE EXPERIMENTS



What do you measure

in which directions:

- k_t for pump-probe experiment
- $2k_t - k_p$ for four wave mixing
- $2k_p - k_t$ for photon echo

INHOMOGENEOUS POLARISATION

Caused by inhomogeneous spatial absorption => $\rho_{ij}(\mathbf{r}_p, \mathbf{r}_t, t) = \langle \psi_j^\dagger(\mathbf{r}_t, t)\psi_i(\mathbf{r}_p, t)\rangle$

center of mass coordinate $\mathbf{R} = (m_i\mathbf{r}_p + m_j\mathbf{r}_t)/(m_i + m_j)$

relative coordinate $\mathbf{r} = \mathbf{r}_p - \mathbf{r}_t$

Wigner distribution: $\rho_{ij}(\mathbf{R}, \mathbf{k}, t) = \frac{1}{V}\int d^3r e^{i\mathbf{k}\cdot\mathbf{r}}\rho_{ij}(\mathbf{R}, \mathbf{r}, t)$

Induced polarisation: $P(\mathbf{R}, t) = \sum_{\mathbf{k}} d_{vc}\rho_{cv}(\mathbf{R}, \mathbf{k}, t) + c.c.$

Another approach: $\rho_{ij}(\mathbf{k}_p, \mathbf{k}_t, t) = \langle a^\dagger_{j,\mathbf{k}_t}(t)\, a_{i,\mathbf{k}_p}(t)\rangle$

Distribution function at spatial coordinate R in a single band:

$$n(\mathbf{R}, \mathbf{k}, t) = \sum_{\mathbf{K}} \rho(\frac{1}{2}\mathbf{K} + \mathbf{k}, -\frac{1}{2}\mathbf{K} + \mathbf{k}, t)e^{i\mathbf{R}\cdot\mathbf{K}}$$

THIN SAMPLES

Calculations in this case from homogeneous fields by adiabatic approximation

Statement: scattered field is proportional to the induced polarisation

Resulting field can be defined from Maxwell equation

$$\frac{\partial^2 \mathcal{E}}{\partial t^2} - \frac{c^2}{n_0^2}\Delta\mathcal{E} = -4\pi\frac{\partial^2 P}{\partial t^2} \simeq 4\pi\omega^2 P$$

Solution: homog. solution + ∫ P · Green function of hom. eq.

≈ weighted spatial average of polarisation

=> E_{transm} ~ P(t, Δt)

ADIABATIC APPROXIMATION

Excitation field

$$\mathcal{E}(t) = \mathcal{E}_p(t) e^{-i\left(\omega_p t - \mathbf{k}_p \cdot \mathbf{r}\right)} + \mathcal{E}_t(t-\tau) e^{-i\left[\omega_t (t-\tau) - \mathbf{k}_t \cdot \mathbf{r}\right]}$$

$$= e^{i\mathbf{k}_p \cdot \mathbf{r}} \left[\mathcal{E}_p(t) e^{-i\omega_p t} + \mathcal{E}_t(t-\tau) e^{-i\omega_t (t-\tau)} e^{i\phi} \right]$$

$\phi = (\mathbf{k}_t - \mathbf{k}_p) \cdot \mathbf{r}$

Induced polarisation

$$P(t,\tau,\phi) = \sum_{\mathbf{k}} d_{vc} \rho_{cv,k}(\phi) + c.c. \propto E_{transm}$$

lattice with lattice vector $\mathbf{k}_p - \mathbf{k}_t$

directions of different orders of diffracted field: $e^{i\mathbf{k}_p \cdot \mathbf{r}} e^{in\phi}$

$n = 1$ into direction $\mathbf{k}_t$

$n = 2$ into direction $2\mathbf{k}_t - \mathbf{k}_p$

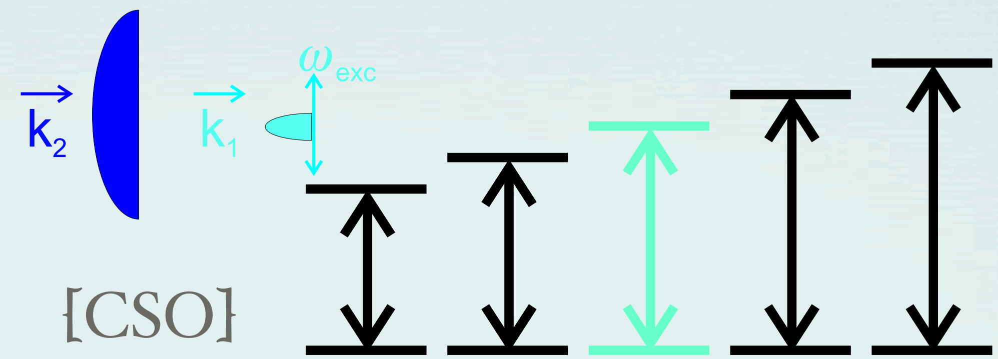
adiabatic approximation:

$$P_n(t,\tau) = \int_0^{2\pi} \frac{d\phi}{2\pi} P(t,\tau,\phi) e^{in\phi}$$

PUMP-PROBE EXPERIMENT

Transmitted light spectrum: $|P_1(\omega,\tau)|^2 \quad (-|P_t^0(\omega)|^2)$

Oscillators with discrete energy distribution:



[CSO]

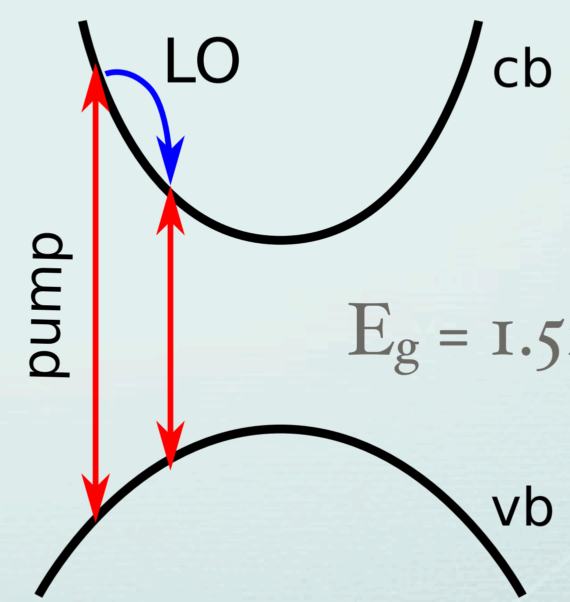
Pump: E_g+150 meV, 1.67 eV;
duration 120 fs;
spectral width 15 meV

Probe: E_g+120 meV, 1.64 eV;
duration 25 fs;
spectral width 70 meV

$\hbar\omega_{LO} = 36$ meV

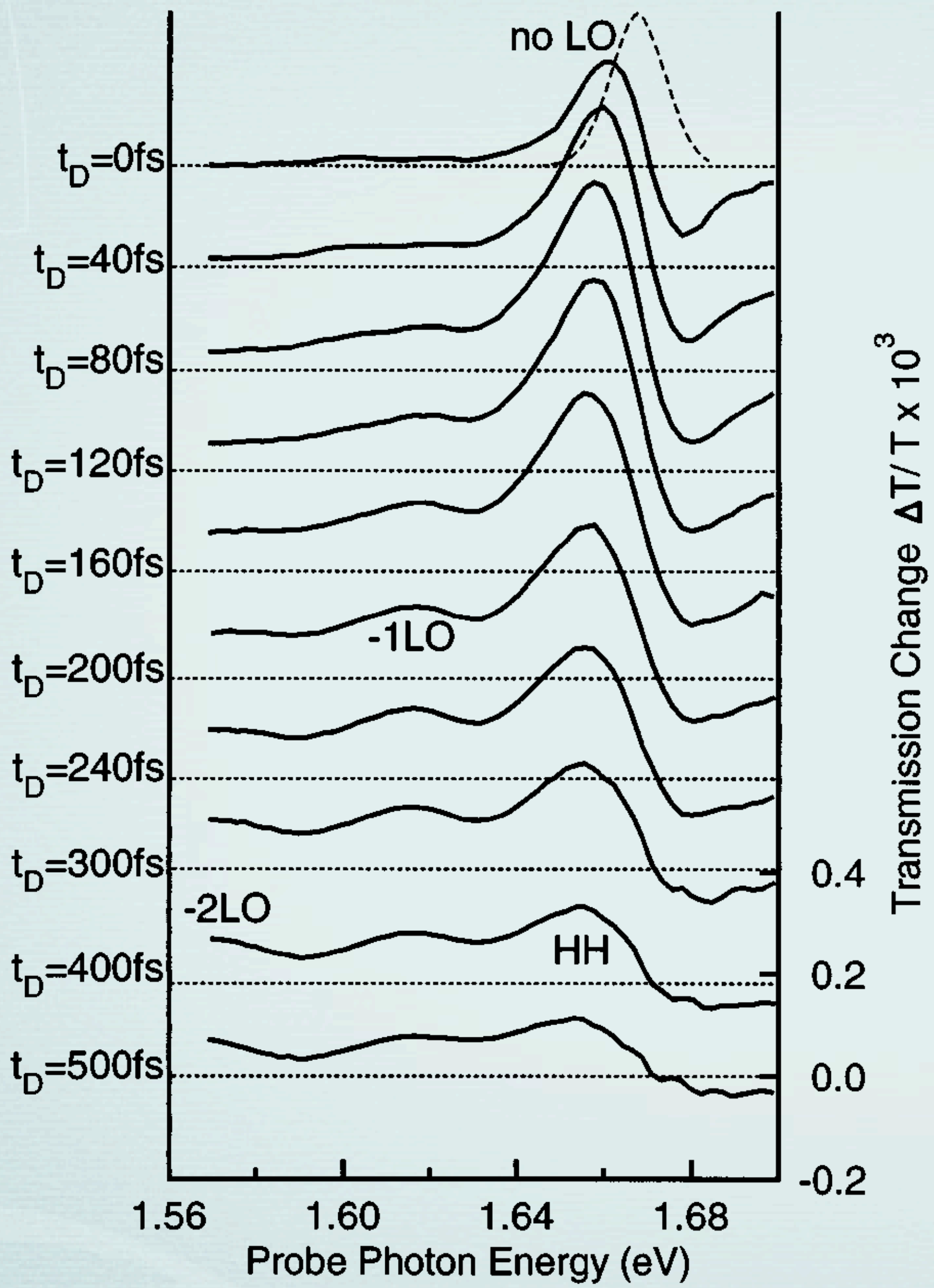
Phonon oscillation time 115 fs

Oscillators with continuum energy distribution:



E_g = 1.52 eV (GaAs)

LEITENSTORFER'S EXPERIMENT

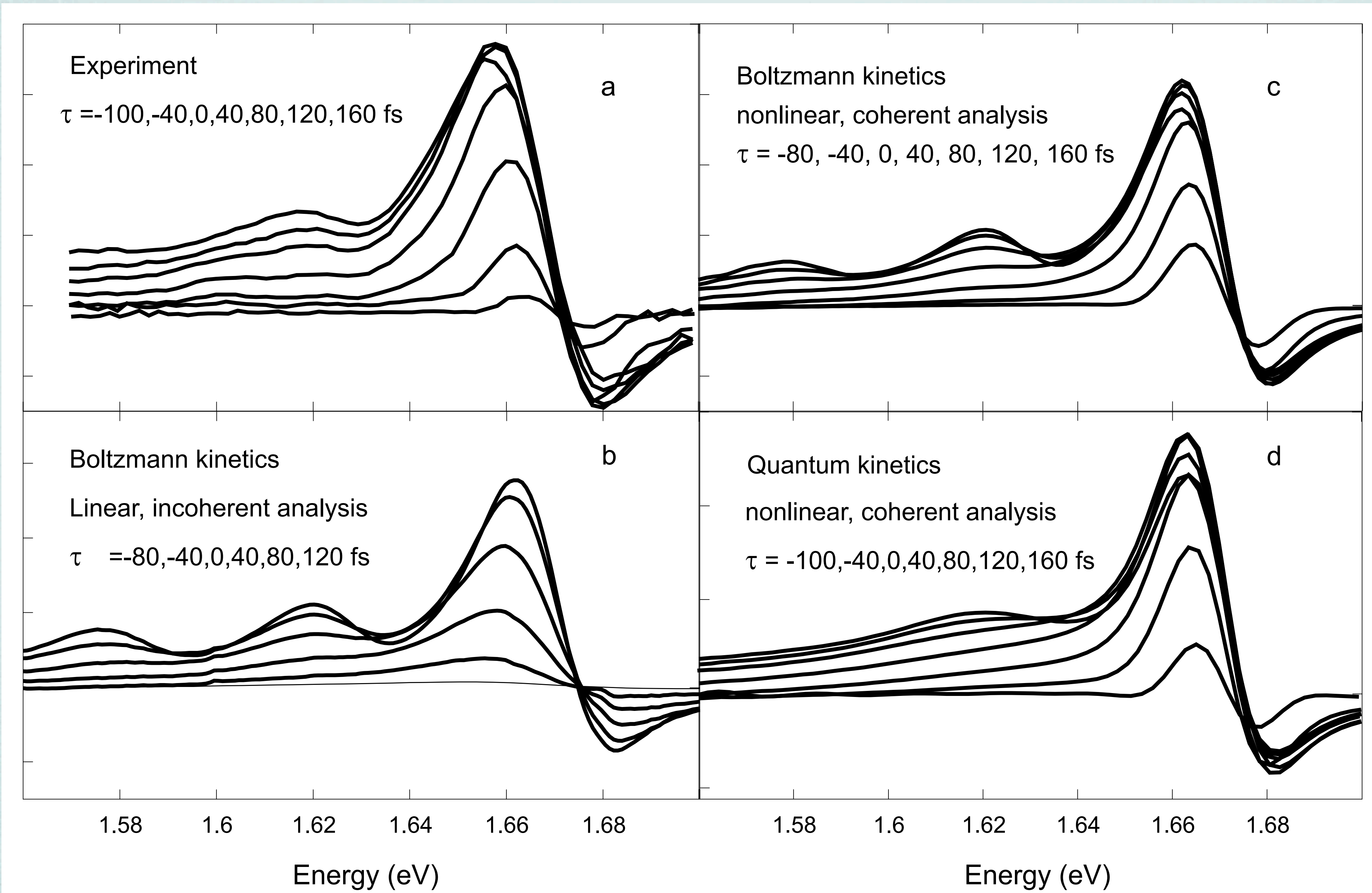


Short time delays (< 100 fs): energy conservation violation

Longer time delays (> 100 fs): memory effects of the system

QUANTUM KINETIC IN ELECTRON-PHONON INTERACTION

Experiment by Fürst, Leitenstorfer *et al.* 1996, theory by Schmenkel *et al.* 1998

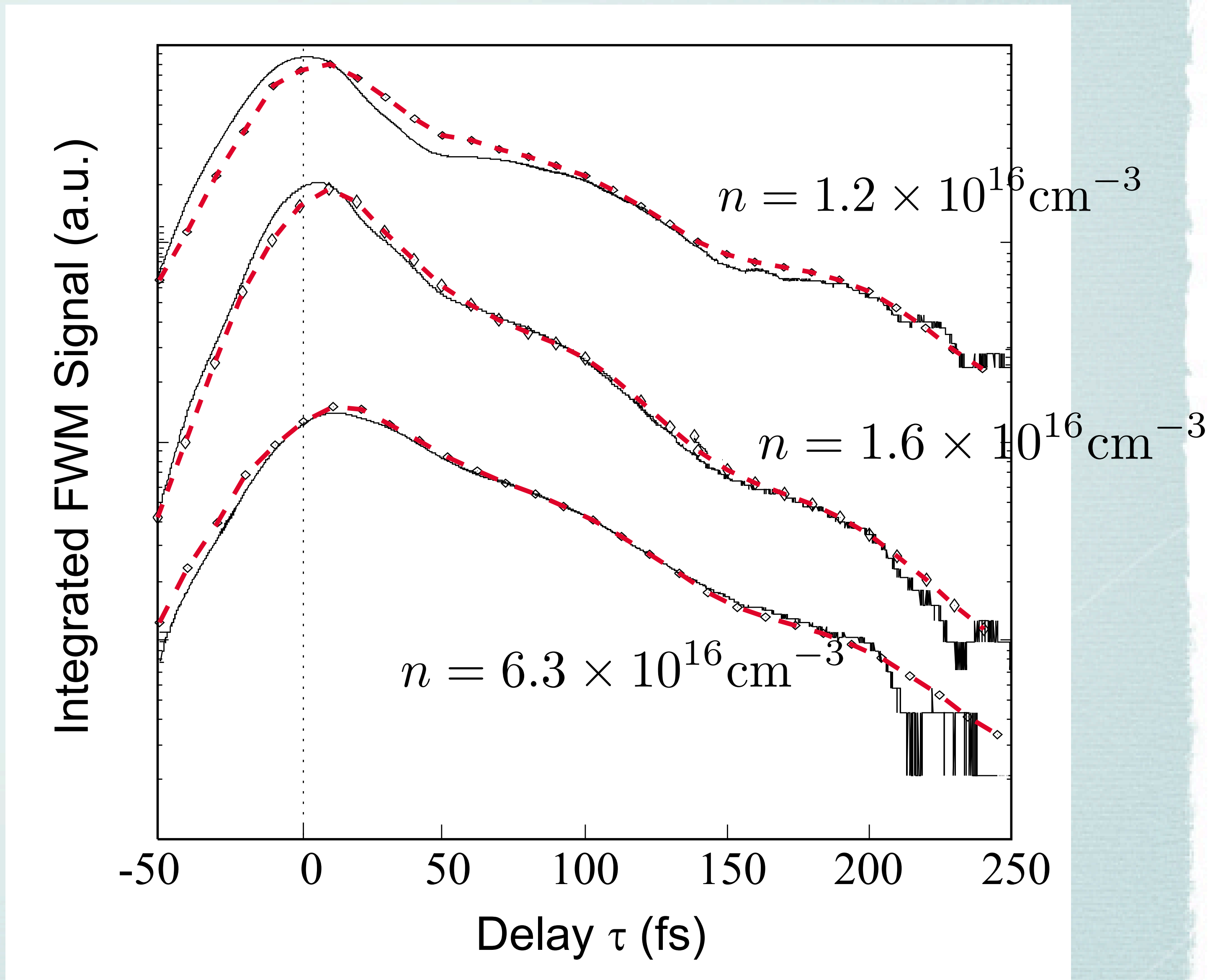


FOUR-WAVE MIXING: LO-PHONONS QUANTUM KINETICS

Time-integrated four-wave mixing signal $\int_{-\infty}^{+\infty} dt |P_2(t,\tau)|^2$

n: density of carriers

Pump & Probe:
sech²-shaped
with FWHM width
14.2 fs & 87 meV



Exponential decay with oscillations with frequency

$$(1 + m_e/m_h)\omega_{LO}$$

Phenomenological damping:

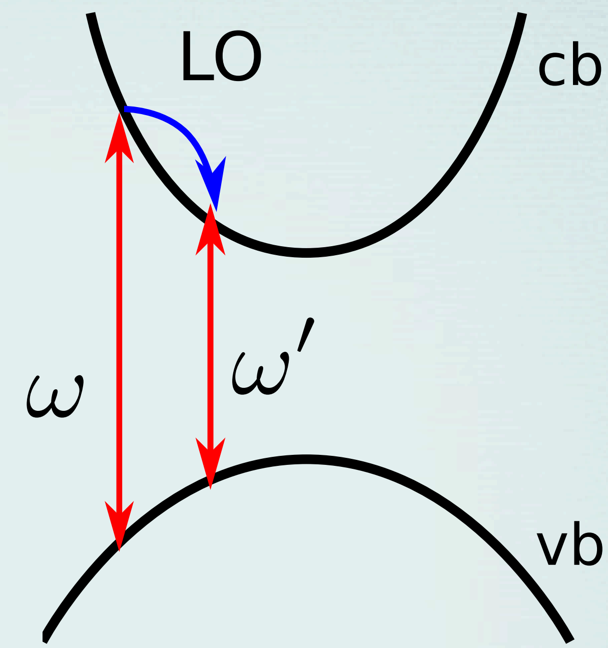
$$\frac{1}{T_2} = \gamma_0 + \gamma_1 n(t)$$

dephasing by phonons (γ_0)

Coulomb scattering ($\gamma_1 n(t)$)

QUANTUM BEATS ASSISTED BY PHONONS

Observed oscillations in decay: „beating of interband-polarisation components with frequency ω und ω' ... connected by coherent LO - phonon scattering" {EPQK}



Resonant band states:

$$\hbar\omega = \hbar^2 k^2/2\mu + E_g'$$

$$\hbar\omega' = \hbar^2 k'^2/2\mu + E_g'$$

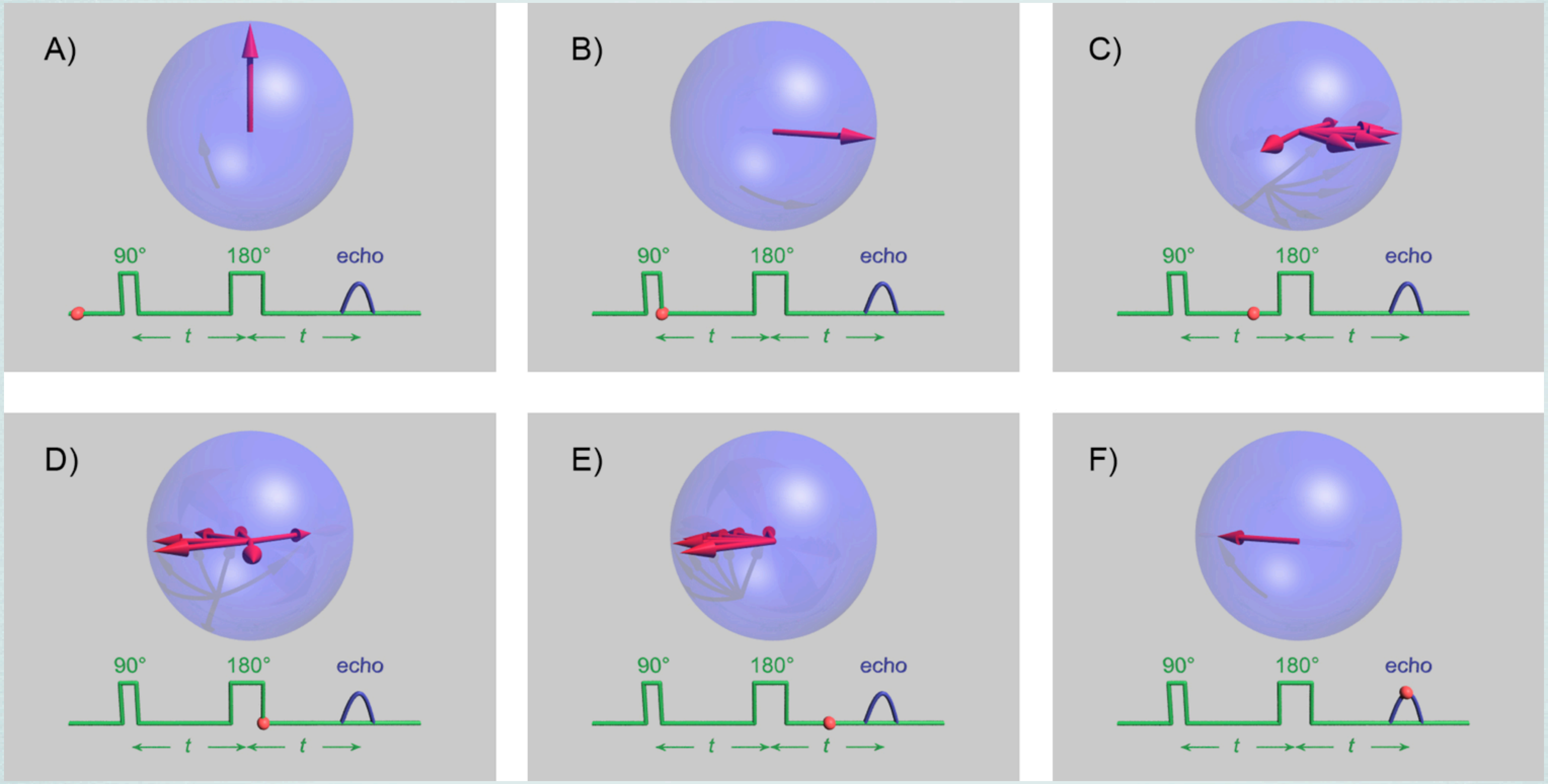
$$\mu = \frac{m_e m_h}{m_e + m_h}$$

renormalised band gap

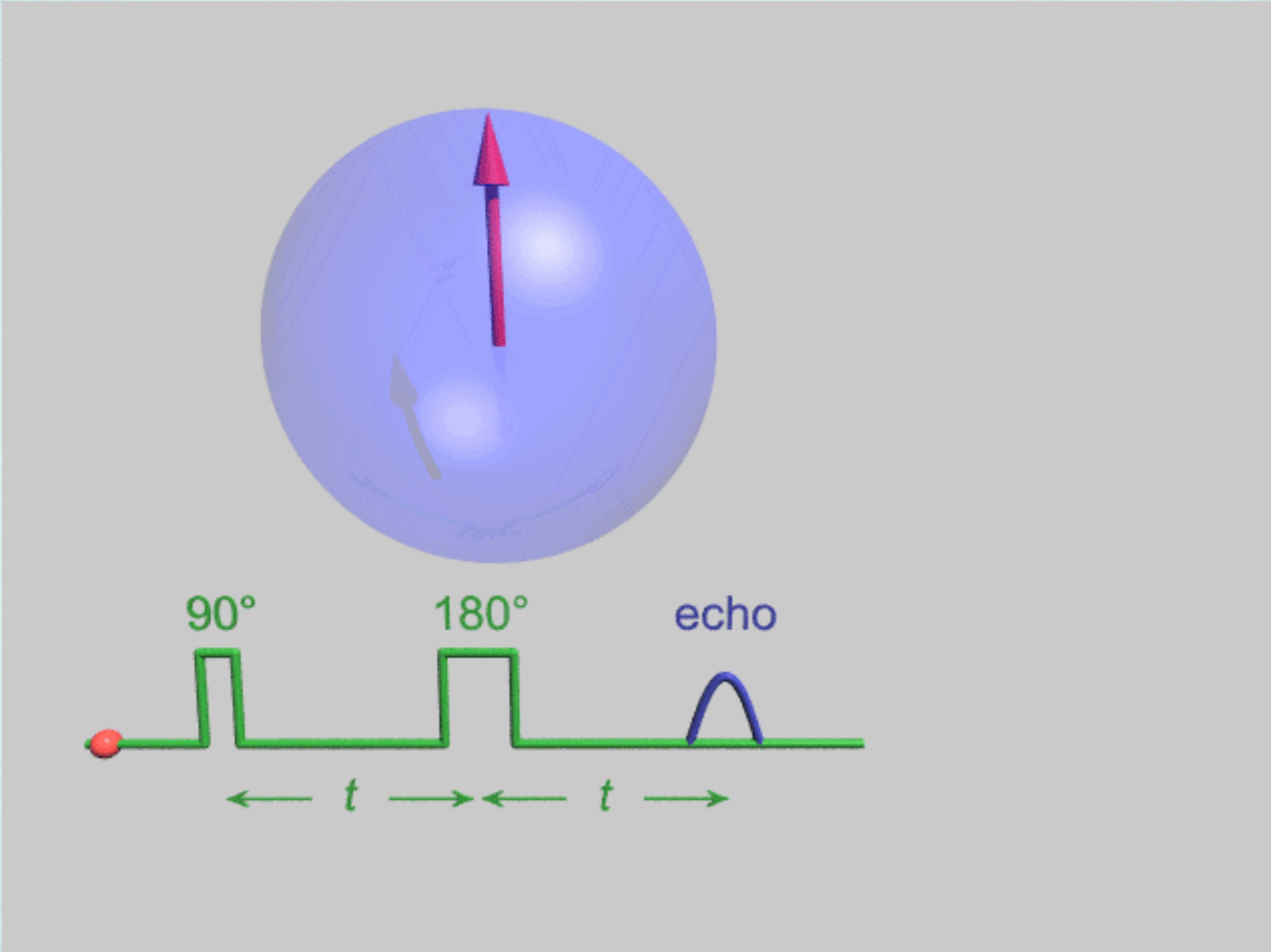
$$\hbar^2(k'^2 - k^2)/2m_e = \hbar\omega_{LO} \implies \omega_{osc} = \omega' - \omega = (1 + m_e/m_h)\omega_{LO}$$

SPIN ECHO (BEFORE PHOTON ECHO)

en.wikipedia.org/wiki/Spin_echo



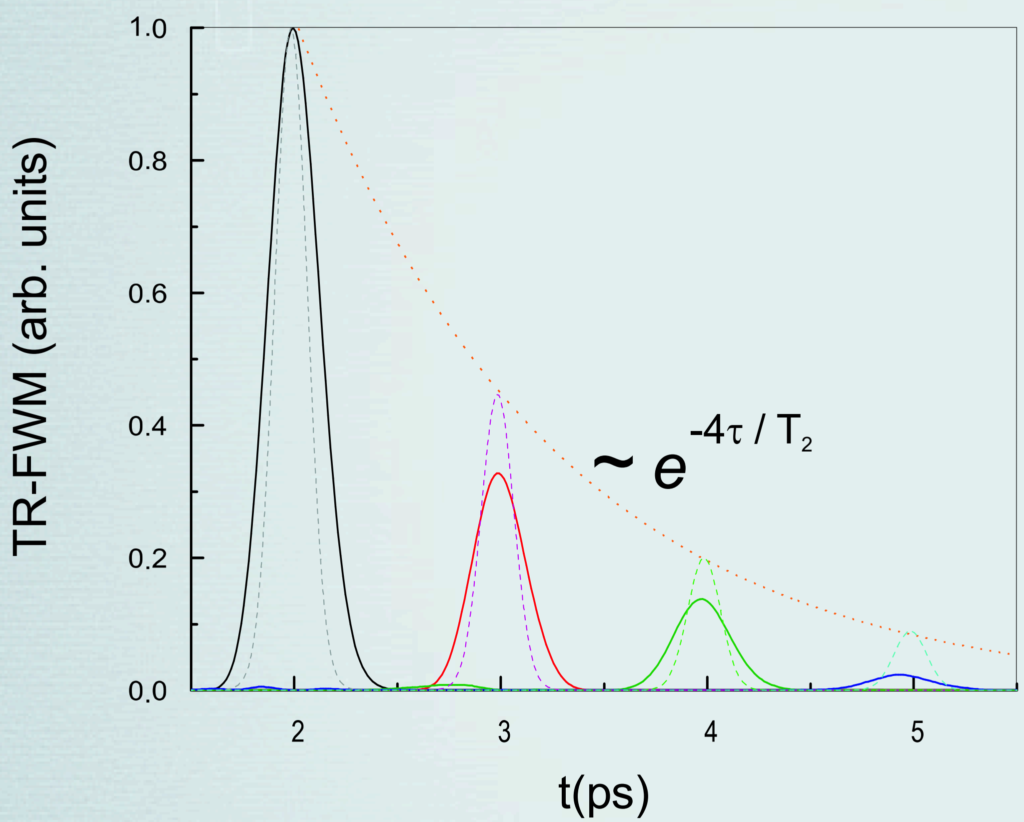
SPIN ECHO SIGNAL



en.wikipedia.org/wiki/Spin_echo

SEMICONDUCTOR PHOTON ECHO

Photon echo from time-resolved 4-wave mixing trace: dashed lines without Coulomb interaction [CSO]



Excitonic photon echo: inhomogeneous broadened system

Instrinstic photon echo: dynamics of interband continuum observed in $2k_p - k_t$ direction

Analytical result: first order of E_t and second order of E_p polarisation:

$$i\frac{dP_k}{dt} = e_k\, P_k - (1-2n_k)\omega_{R,k}$$

$$\frac{dn_k}{dt} = i(\omega_{R,k}P_k^* - \omega_{R,k}^* P_k)$$

Use conservation law $n_k = \frac{1}{2}(1 \pm \sqrt{1-4|P_k|^2})$ →

SOLVING THE BLOCH EQUATIONS

Interband polarisation:

$$i\hbar\frac{d}{dt}P_k = \hbar\epsilon_k\,P_k - \sum_{k'} V_{k'}\,P_{k+k'} - 2\sum_{k'} V_{k'}\,|P_{k+k'}|^2 P_k + 2\sum_{k'} V_{k'}\,P_{k+k'}\,|P_k|^2 - d_{cv}\,E(1-2|P_k|^2)$$

Solution procedure can be shown on simplified equation:

$$i\frac{dP}{dt} = \epsilon\,P + E + a|P|^2 E + b|P|^2\,P$$

Solving for first order of E_t:

$$i\,\frac{dP}{dt} = \epsilon\,P + E_t$$

Polarisation for time t_o, when E_t is gone and E_p not arrived yet

$$P(t_0) = -i\,e^{-i\epsilon t_0}\,E_t(\epsilon) \qquad E_t(\epsilon) = \int_{-\infty}^{t_0} dt\,E_t(t)\,e^{i\epsilon t}$$

FIELD IN DIRECTION 2KP-KT

Polarisation after arriving of E_p:

$$P^{(0)}(t) = -i\, e^{-i\epsilon t} \left[E_t(\epsilon) + \int_{t_0}^{t} dt'\, E_p(t')\, e^{i\epsilon t'} \right]$$

By iteration:

$$i\, \frac{dP^{(1)}}{dt} = \epsilon\, P^{(1)} + E_p + a|P^{(0)}|^2 E_p + b|P^{(0)}|^2 P^{(0)}$$

Direction of interest: $2\mathbf{k}_p - \mathbf{k}_t$

Considering only term $\propto E_p\, E_p\, E_t^*$:

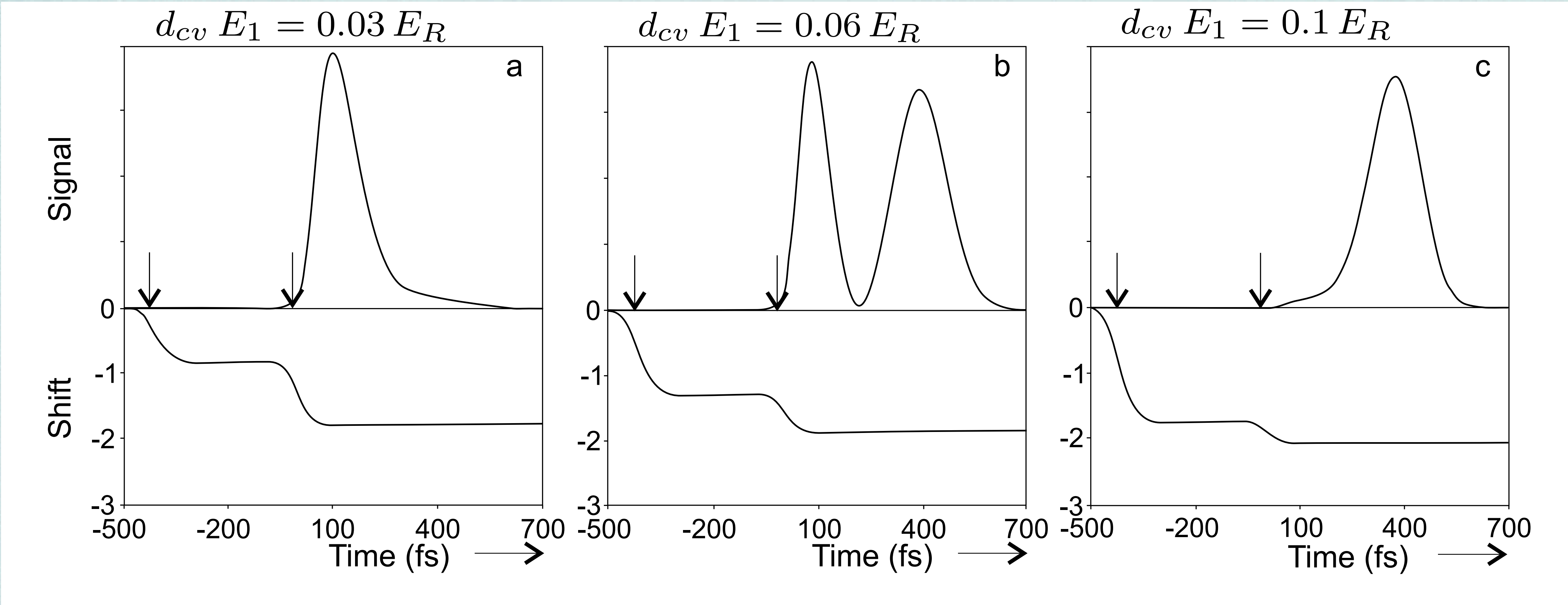
$$P(t) \frac{\hbar^3}{|d_{cv}|^2 d_{cv}} = -2i \sum_{\lambda} E_t^*(\lambda)\, e^{-i\epsilon_\lambda t} \left[\int_{t_0}^{t} dt'\, E_p(t')\, e^{i\epsilon_\lambda t'} \right]^2 - 2 \sum_{\lambda\lambda'} V_{\lambda-\lambda'}\, E_t^*(\lambda')\, e^{-i\epsilon_\lambda t} \int_{t_0}^{t} dt' \left[1 - e^{i(\epsilon_\lambda - \epsilon_{\lambda'})(t-t')} \right]$$
$$\times \int_{t_0}^{t'} dt''\, E_p(t'')\, e^{i\epsilon_\lambda t''} \int_{t_0}^{t'} dt'''\, E_p(t''') e^{i\epsilon_{\lambda'} t'''}$$

Assuming $E_t(t) = E_t\, \delta(t+\tau)$

$E_p(t) = E_p\, \delta(t)$

First term of P(t): $P_{(1)}(t) \propto E_t^* E_p^2 \sum_{\lambda} e^{-i\epsilon_\lambda (t-\tau)} \propto E_t^*\, E_p^2\, \delta(t-\tau)$

PHOTON ECHO OF CONTINUUM STATES



$d_{cv}\,E_2 = 0.1\,E_R$

$$\text{Shift} = \frac{E_G - E_G^0}{E_R}$$

E_G : renormalised band edge

E_G^0 : unrenormalised band edge

E_R : exciton binding energy (16meV in CdSe)

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