Optical Stark Effect

Michael Körber

Optical Properties of Semiconductors
Universität Konstanz

June 14, 2011

Contents

Repetition

Optical Stark effect of a two-level atom in an electric field

$$\mathcal{E}(t) = \frac{1}{2}\mathcal{E}(\omega)\left(e^{-i\omega t} + \text{c.c.}\right)$$

Interaction with electric field

$$\mathcal{H}_{int} = -d\mathcal{E}(t)$$

Evaluation of time-dependent coefficients a_n in

$$\psi(\boldsymbol{r}, t) = \sum_n a_n(t) e^{-i\epsilon_n t}\psi_n(\boldsymbol{r})$$

Found coupled differential equations;

$$i\hbar\frac{\mathrm{d}a_1}{\mathrm{d}t} = -\mathcal{E}(t)e^{-i\epsilon_{21}t}d_{12}a_2$$
$$i\hbar\frac{\mathrm{d}a_2}{\mathrm{d}t} = -\mathcal{E}(t)e^{-i\epsilon_{21}t}d_{21}a_1$$

Repetition

Only resonant part $\omega - \epsilon_{21}$ (rotating wave approximation).

Resonance ($\omega = \epsilon_{21}$):

$$\frac{\mathrm{d}^2 a_2}{\mathrm{d}t^2} = -\frac{\omega_R^2}{4}a_2 \qquad \text{with} \qquad \omega_R = \frac{|d_{21}\mathcal{E}|}{\hbar}$$

Splitting of energy niveaus:

$$\epsilon_i \to \epsilon_i \pm \frac{\omega_R}{2}$$

With finite detuning $\nu = \epsilon_{21} - \omega$:

$$\epsilon_2 \to \epsilon_2 - \frac{\nu}{2} \pm \frac{1}{2}\sqrt{\nu^2 + \omega_R^2}$$
$$\epsilon_1 \to \epsilon_1 + \frac{\nu}{2} \pm \frac{1}{2}\sqrt{\nu^2 + \omega_R^2}$$

Repetition

Three possible transitions:

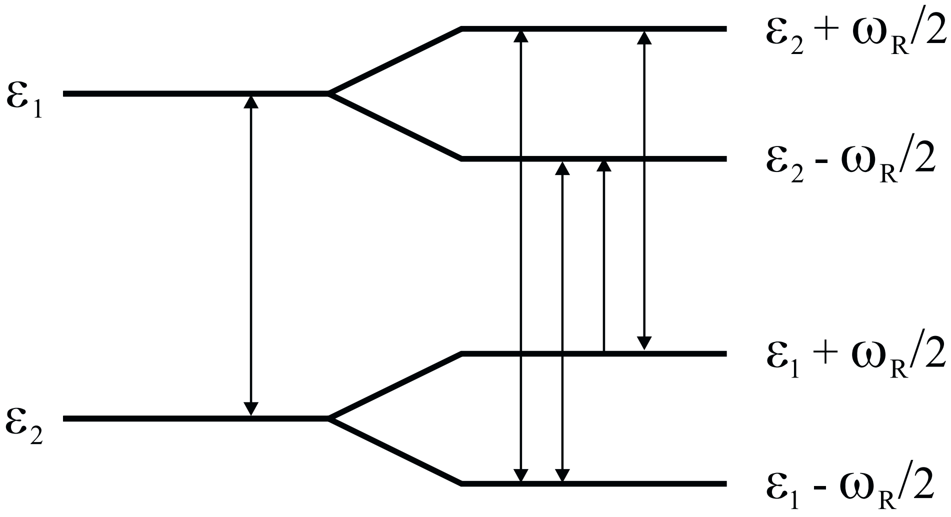


Figure: Splitting of energy levels in a two-level atom in a resonant electric field [Haug].

Two-level atom in 2. quantization

Two-level Hamiltonian:

$$\mathcal{H} = \sum_{j=1,2} \hbar\epsilon_j a_j^\dagger a_j - \left[d_{21}\mathcal{E}(t)a_2^\dagger a_1 + \text{h.c.}\right] \tag{1}$$

with coherent light field $\mathcal{E}(t) = \mathcal{E}_p e^{-i\omega_p t}$.

Time-dependency via Heisenberg equations.

Polarization $P = \left\langle a_1^\dagger a_2 \right\rangle$:

$$\frac{\mathrm{d}P}{\mathrm{d}t} = -i\epsilon P + \frac{id_{21}}{\hbar}\mathcal{E}(t)\,(1-2n) \tag{2}$$

Density of upper state $n = \left\langle a_2^\dagger a_2 \right\rangle = 1 - \left\langle a_1^\dagger a_1 \right\rangle$:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \frac{i}{\hbar}\left(d_{21}\mathcal{E}(t)P^* - \text{h.c.}\right) \tag{3}$$

Two-level atom in 2. quantization

Neglected Damping $\Rightarrow$ completely coherent process

Conserved quantity:

$$K = (1 - 2n)^2 + 4\,|P|^2$$

with initial conditions $n = 0$ and $P = 0$:

$$n = \frac{1}{2}\left(1 \pm \sqrt{1 - 4\,|P|^2}\right) \tag{4}$$

density fully described by polarization
(only true for virtual excitations)

Two-level atom in 2. quantization

Fourier Transformation

$$\widetilde{P}(\omega) = \int \mathrm{d}t e^{i\omega t} P(t)$$

Set of equations in frequency-space

$$\begin{aligned}
(\omega - \epsilon)\,\widetilde{P} &= -\frac{d_{21}}{\hbar}\mathcal{E}_p\left(\delta(\omega - \omega_p) - 2\widetilde{n}(\omega - \omega_p)\right)\\
(-\omega - \epsilon)\,\widetilde{P}^* &= -\frac{d_{21}^*}{\hbar}\mathcal{E}_p\left(\delta(\omega + \omega_p) - 2\widetilde{n}(\omega + \omega_p)\right)\\
\omega\widetilde{n} &= \frac{1}{\hbar}\left(d_{21}^*\mathcal{E}_p\widetilde{P}(\omega + \omega_p) - d_{21}\mathcal{E}_p\widetilde{P}^*(\omega - \omega_p)\right)
\end{aligned}$$

Solving homogeneous part:

$$(\omega - \omega_p)\left[(\omega - \epsilon)(-\omega - \epsilon + 2\omega_p) + 4\omega_R^2\right] = 0 \tag{5}$$

Two-level atom in 2. quantization

$$(\omega - \omega_p)\left[(\omega - \epsilon)(-\omega - \epsilon + 2\omega_p) + 4\omega_R^2\right] = 0 \quad (5)$$

Resonant Part ($\omega_p = \epsilon$):

$$\omega = \begin{cases} \epsilon & +2\omega_R \\ \epsilon & \\ \epsilon & -2\omega_R \end{cases} \quad (6)$$

Finite Detuning $\nu = \epsilon - \omega_p$:

$$\omega = \begin{cases} \epsilon - \nu & +\sqrt{\nu^2 + 4\omega_R^2} \\ \epsilon - \nu & \\ \epsilon - \nu & -\sqrt{\nu^2 + 4\omega_R^2} \end{cases} \quad (7)$$

„Mollow triplet"

Excitonic Optical Stark Effect
Introduction

- Optical Relaxation has short characteristic times in semiconductors
- Examination of coherent effects needs femtosecond laser pulses
- Bound states as excitons have longer characteristic times (picoseconds)
- First obeserved in 1980's

Excitonic Optical Stark Effect
Configuration

Two time-resolved pulses:

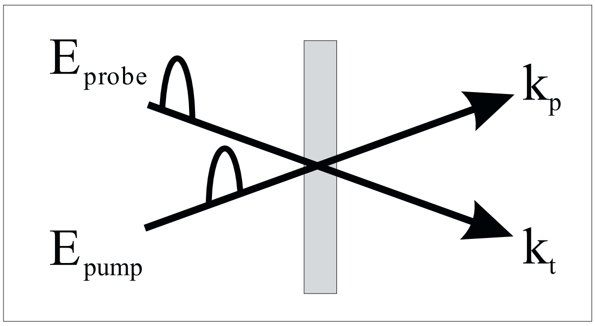


Figure: Pump-Probe Experiment [Haug].

- Strong detuned pump beam
- Short probe beam at resonance
- Measurement of change in absorption $\delta\alpha = \alpha_p - \alpha_{np}$

Excitonic Optical Stark Effect
Quasi-stationary approximation

Consider only the pump beam
Later: probe beam as perturbation

Semiconductor Bloch equations:

$$\frac{\mathrm{d}P_k}{\mathrm{d}t} = -ie_k P_k - (1-2n_k)\,\omega_{R,k} \tag{8}$$

$$\frac{\mathrm{d}n_k}{\mathrm{d}t} = i\left(\omega_{R_k} P_k^* - \omega_{R,k}^* P_k\right) \tag{9}$$

with

$$\hbar e_k = \hbar\left(e_{e,k} + e_{h,k}\right) = E_g + \frac{\hbar^2 k^2}{2m} - 2\sum_{k'} V_{k-k'} n_{k'}$$

$$\omega_{R,k} = \frac{1}{\hbar}\left(d_{cv}\mathcal{E}(t) + \sum_{q\neq k} V_{|k-q|} P_q\right)$$

Excitonic Optical Stark Effect
Quasi-stationary approximation

Analogically to two-level atom (same conserved quantity K):

$$n_k = \frac{1}{2}\left(1 \pm \sqrt{1 - 4\left|P_k\right|^2}\right) \tag{10}$$

Solving Bloch equation adiabatically with $\mathcal{E}(t) = E_p \exp(-i\omega_p t)$ and

$$P_k(t) = p_k(t)e^{-i\omega_p t} \approx p_k e^{-i\omega_p t}.$$

Adiabatic solution:

$$P_k = \frac{(1-2n_k)\omega_{R,k}}{e_k - \omega_p} \tag{11}$$

Excitonic Optical Stark Effect
Quasi-stationary approximation

Add weak probe beam $\mathcal{E}_t \exp(-i\omega_t t)$

Small change in polarization $P_k \to P_k + \delta P_k$:

$$i\frac{\mathrm{d}}{\mathrm{d}t}\delta P_k = \delta e_k P_k + e_k \delta P_k + 2\delta n_k \omega_{R,k} - (1 - 2n_k)\,\delta\omega_{R,k} \tag{12}$$

with

$$\delta e_k = -\frac{2}{\hbar}\sum_{k'} V_{k-k'}\delta n_{k'}$$

$$\delta n_k = \frac{P_k \delta P_k^* + P_k^* \delta P_k}{1 - 2n_k}$$

$$\delta\omega_{R,k} = \frac{d_{cv}}{\hbar}\mathcal{E}_t e^{-i\omega_t t} + \frac{1}{\hbar}\sum_{k'} V_{k-k'}\delta P_{k'}$$

Excitonic Optical Stark Effect
Quasi-stationary approximation

$$i\frac{\mathrm{d}}{\mathrm{d}t}\delta P_k = \delta e_k P_k + e_k \delta P_k + 2\delta n_k \omega_{R,k} - (1-2n_k)\,\delta\omega_{R,k} \tag{12}$$

Eliminate time dependence of pump field ($P_k = p_k e^{-i\omega_p t}$)

$$i\frac{\mathrm{d}}{\mathrm{d}t}\delta p_k = \delta e_k p_k + (e_k - \omega_p)\,\delta p_k + 2\delta n_k \omega_{R,k} - (1-2n_k)\,\delta\omega_{R,k} \tag{13}$$

Redefined change in Rabi frequency

$$\delta\omega_{R,k} = \frac{d_{cv}\mathcal{E}_t}{\hbar} e^{i\Delta t} + \frac{1}{\hbar}\sum_{k'} V_{k-k'}\delta p_{k'} \tag{14}$$

with $\Delta = \omega_p - \omega_t$.

Excitonic Optical Stark Effect
Quasi-stationary approximation

Solve with

$$\delta p_k = \delta p_k^+ e^{i\Delta t} + \delta p_k^- e^{-i\Delta t} \tag{15}$$

(P_k and n_k must be known)

δp_k^+ contributes resonant part.
(Only numerical solvable)

$\rightarrow$ susceptibility and absorption spectrum

Excitonic Optical Stark Effect
Quasi-stationary approximation

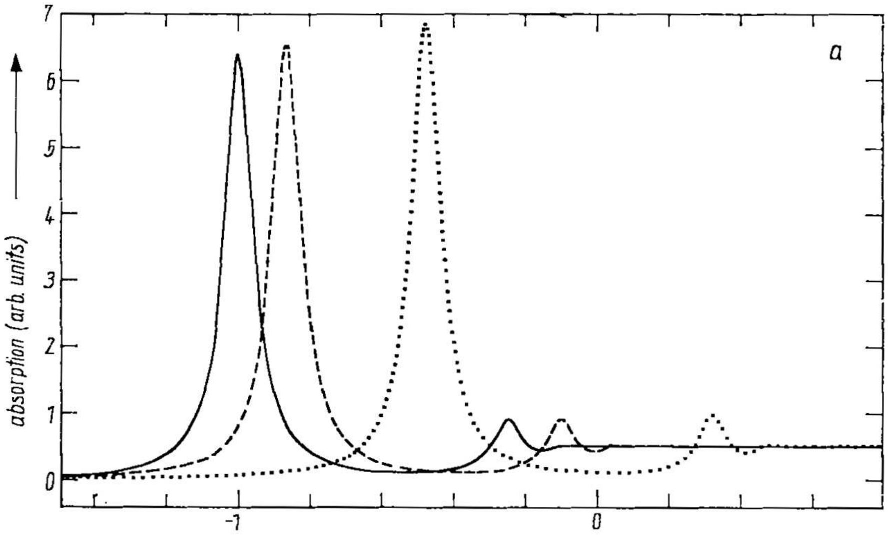


Figure: Calculated absorption spectrum versus normalized detuning of $(\hbar\omega_t - E_g)/E_0$ for a pump detuning $(E_g - \hbar\omega_p)/E_0 = 10$ with $I_p = 0,\ 7.5,\ 30\ \mathrm{MW/cm^2}$ from left to right [Ell].

Excitonic Optical Stark Effect
Dynamic results

Now: Take dynamic effects into account.

$$\mathcal{E}(r,t) = \mathcal{E}_p(t)e^{-i(\boldsymbol{k}_p\cdot\boldsymbol{r}+\Omega t)} + \mathcal{E}_t(t)e^{-(\boldsymbol{k}_t\cdot\boldsymbol{r}+\Omega t)}$$

Bloch equations in linear order (ignore terms $\mathcal{O}(n \cdot P)$):

$$\hbar\left[i\frac{\partial}{\partial t} - \epsilon_k\right]P_{\boldsymbol{k}} = (2n_{\boldsymbol{k}} - 1)d_{cv}\mathcal{E}(t) - \sum_{\boldsymbol{q}\neq\boldsymbol{k}} V_{|\boldsymbol{k}-\boldsymbol{q}|}P_{\boldsymbol{q}} \tag{16}$$

$$\hbar\frac{\partial}{\partial t}n_{\boldsymbol{k}} = i\left[d_{cv}\mathcal{E}(t)P^*_{\boldsymbol{k}} - \text{c.c.}\right] \tag{17}$$

Transformation to real space:

$$i\hbar\frac{\partial}{\partial t}P(\boldsymbol{r}) = \mathcal{H}_{eh}P(\boldsymbol{r}) + d_{cv}\mathcal{E}(t)\left[2n(\boldsymbol{r}) - \delta(r)\right] \tag{18}$$

$$\hbar\frac{\partial}{\partial t}n(\boldsymbol{r}) = i\left[d_{cv}\mathcal{E}(t)P^*(-\boldsymbol{r}) - \text{c.c.}\right] \tag{19}$$

Wannier Hamiltonian:

$$\mathcal{H}_{eh} = \epsilon_g - \frac{\hbar^2\nabla^2}{2m_r} - V(r) \tag{20}$$

Find equations in orders of the Wannier equations

$$\int \mathrm{d}\boldsymbol{r}\ \psi_\lambda(\boldsymbol{r}) i\hbar\frac{\partial}{\partial t}P(\boldsymbol{r}) = \int \mathrm{d}\boldsymbol{r}\ \psi_\lambda(\boldsymbol{r})\mathcal{H}_{eh}P(\boldsymbol{r}) + \int \mathrm{d}\boldsymbol{r}\ \psi_\lambda(\boldsymbol{r}) d_{cv}\mathcal{E}(t)\left[2n(\boldsymbol{r}) - \delta(r)\right]$$

$$\int \mathrm{d}\boldsymbol{r}\ \psi_\lambda(\boldsymbol{r})\hbar\frac{\partial}{\partial t}n(\boldsymbol{r}) = \int \mathrm{d}\boldsymbol{r}\ \psi_\lambda(\boldsymbol{r}) i\left[d_{cv}\mathcal{E}(t)P^*(-\boldsymbol{r}) - \text{c.c.}\right]$$

$\Rightarrow$ equations closed in λ:

$$i\hbar\frac{\partial}{\partial t}P_\lambda = \hbar\epsilon_\lambda P_\lambda + d_{cv}\mathcal{E}(t)\left[2n_\lambda - \psi_\lambda(r=0)\right] \tag{21}$$

$$\hbar\frac{\partial}{\partial t}n_\lambda = id_{cv}\mathcal{E}(t)P_\lambda^* - id_{cv}\mathcal{E}(t)P_\lambda \tag{22}$$

Eliminate time-dependency of electric field and reduce to s-functions by $P_\lambda = \psi_\lambda(r=0)e^{-i\Omega t}p_\lambda$.
Introduce damping γ, Γ and $w_\lambda = \psi_\lambda(r=0)(1-2n_\lambda)$:

$$\frac{\partial}{\partial t}p_\lambda = [i(\epsilon_\lambda - \Omega) + \gamma]\, p_\lambda + i\frac{d_{cv}}{\hbar}\left[\mathcal{E}_p(t)e^{-i\boldsymbol{k}_p\cdot\boldsymbol{r}} + \mathcal{E}_t(t)e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}}\right] w_\lambda \tag{23}$$

$$\frac{\partial}{\partial t}w_\lambda = -\Gamma(w_\lambda - 1) - i\frac{2d_{cv}}{\hbar}\left[\mathcal{E}_p(t)e^{-i\boldsymbol{k}_p\cdot\boldsymbol{r}} + \mathcal{E}_t(t)e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}}\right] p_\lambda^* + i\frac{2d_{cv}^*}{\hbar}\left[\mathcal{E}_p^*(t)e^{i\boldsymbol{k}_p\cdot\boldsymbol{r}} + \mathcal{E}_t^*(t)e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}}\right] p_\lambda \tag{24}$$

Short probe pulse:

$$\mathcal{E}_t(t) \simeq \mathcal{E}_t\delta(t - t_t)$$

Measurement in probe beam direction:

$$\propto e^{i\boldsymbol{k}_t\cdot\boldsymbol{r}}$$

Now:
Solve p_λ for this setting.

Weak probe pulse $\Rightarrow$ First order in $\mathcal{E}_t$
Only first order in p_λ $\Rightarrow$ Need second order in $\mathcal{E}_p$

Integration over equation of motion yields:

$$p_\lambda(t) \simeq i\frac{d_{cv}}{\hbar}\mathcal{E}_t e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}} e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t-t_t)} w_\lambda(t_{t_-})\Theta(t-t_t) \\ \underbrace{+ i\frac{d_{cv}}{\hbar} e^{-i\boldsymbol{k}_p\cdot\boldsymbol{r}} \int_{-\infty}^{t} \mathrm{d}t' e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t-t_t)} \mathcal{E}_p(t') w_\lambda(t')}_{p_\lambda(t<t_t)} \quad (25)$$

w_λ contains a part $\propto e^{i\boldsymbol{k}_t\cdot\boldsymbol{r}}$ for $t_{t_-} < t < t_{t_+}$.

Polarization shows terms $\propto e^{i(\boldsymbol{k}_t-\boldsymbol{k}_p)\cdot\boldsymbol{r}}$.

$\Rightarrow$ scattering of pump beam in probe direction possible.

Same analysis for $w_\lambda(t)$ and recursive inserting provides:

$$p_\lambda(t) \simeq i\frac{d_{cv}}{\hbar}\mathcal{E}_t e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}} e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t-t_t)} w_\lambda(t_{t_-})\Theta(t-t_t) \\ + 2\frac{d_{cv}^2}{\hbar^2}\mathcal{E}_t e^{-i(\boldsymbol{k}_t+\boldsymbol{k}_p)\cdot\boldsymbol{r}} p_\lambda^*(t_{t_-}) \\ \times \int_{t_t}^{t} \mathrm{d}t' e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t-t_t)} e^{-\Gamma(t'-t_t)} \mathcal{E}_p(t')\Theta(t-t_t) \\ - i\frac{2d_{cv}}{\hbar}\frac{|d_{cv}|^2}{\hbar^2}\mathcal{E}_t e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}} \int_{t_t}^{t} \mathrm{d}t' e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t-t')} \mathcal{E}_p(t') \\ \times \int_{t_t}^{t'} \mathrm{d}t'' e^{-\Gamma(t'-t'')} \mathcal{E}_p^*(t'') e^{-[i(\epsilon_\lambda-\Omega)+\gamma](t''-t_t)} \Theta(t-t_t) \quad (26)$$

Excitonic Optical Stark Effect
Dynamic results

Transformation to Fourier-space:

$$\mathcal{P}_\lambda(\omega) = \int \mathrm{d}t\; e^{i\omega t}\mathcal{P}_\lambda(t) = \int \mathrm{d}t\; e^{i(\omega-\Omega)t} p_\lambda(t)$$
$$= \int_0^\infty \mathrm{d}t\; e^{i(\omega-\Omega)t} p_\lambda(t+t_t) e^{-i(\omega-\Omega)t_t}$$
$$\mathcal{E}_t(\omega) = \int \mathrm{d}t\; e^{i\omega t}\mathcal{E}_t(t) \simeq \mathcal{E}_t e^{i(\omega-\Omega)t_t} e^{-i\boldsymbol{k}_t\cdot\boldsymbol{r}}$$

Calculate susceptibility:

$$\chi_\lambda(\omega) = \frac{\mathcal{P}_\lambda(\omega)}{\mathcal{E}_t(\omega)} \tag{27}$$

Excitonic Optical Stark Effect
Dynamic results

Slow varying pump beam:

$$\delta\chi_\lambda(\omega) \simeq 2\frac{d_{cv}}{\hbar}\frac{|d_{cv}|^2}{\hbar^2}\frac{1}{\omega-\Omega}\int_0^\infty \mathrm{d}t\; \frac{|E_p(t+t_t)|^2}{\gamma - i(\omega-\epsilon_\lambda)} e^{-[i(\epsilon_\lambda-\omega)+\gamma]t} \tag{28}$$

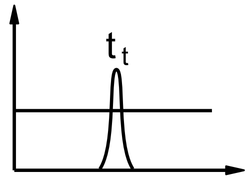
Differential Absorption:

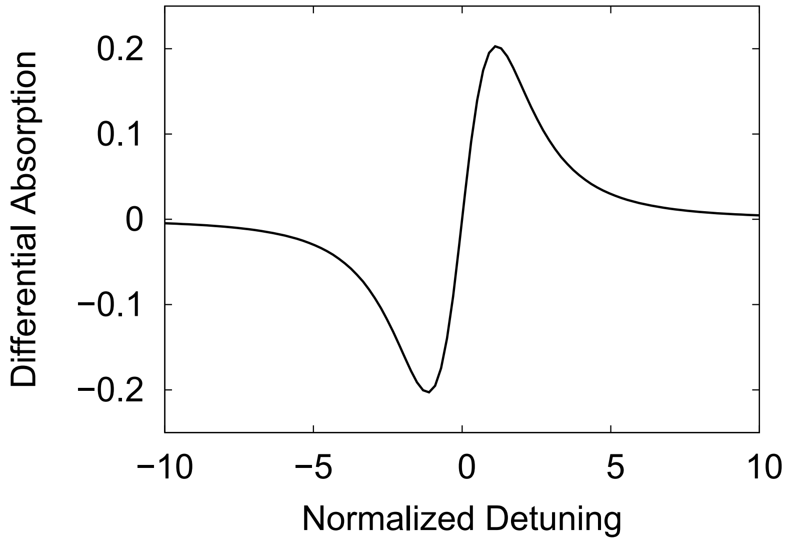
$$\delta\alpha(\omega) \simeq \frac{|d_{cv}|^4}{\hbar^4}\frac{2}{\Omega-\epsilon_\lambda}\Im\left[\int_0^\infty \mathrm{d}t\; \frac{|E_p(t+t_t)|^2}{\gamma - i(\omega-\epsilon_\lambda)} e^{-[i(\epsilon_\lambda-\omega)+\gamma]t}\right] \tag{29}$$

Excitonic Optical Stark Effect
Dynamic results

Constant beam $\mathcal{E}_p(t) = \text{const.}$:

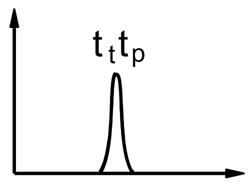
$$\delta\alpha(\omega) \propto -\frac{|\mathcal{E}_p|^2}{\epsilon_\lambda - \Omega_p} \frac{2\gamma(\epsilon_\lambda - \omega)}{[(\epsilon_\lambda - \omega)^2 + \gamma^2]^2} \qquad (30)$$





Excitonic Optical Stark Effect
Dynamic results

120 fs Gaussian pump pulse $\mathcal{E}_p(t) = \mathcal{E}_0 e^{\left(-\frac{(t-t_p)^2}{2\sigma^2}\right)}$:



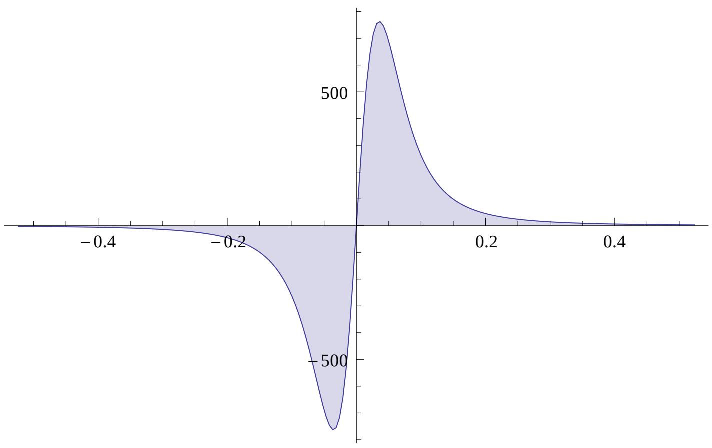


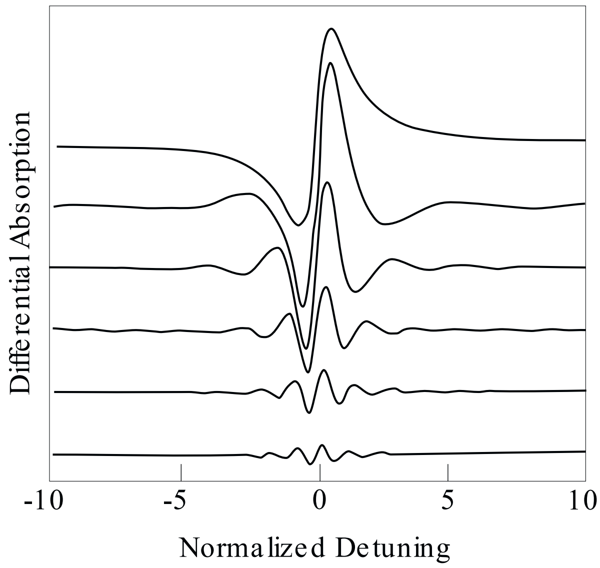
Figure: Differential absorption versus normalized detuning $(\omega - \epsilon_\lambda)/\sigma$ for $t_t = t_p$.

Excitonic Optical Stark Effect
Dynamic results

Short pump pulse:
→ Consider whole susceptibility

$$\chi_\lambda(\omega) \simeq i\frac{d_{cv}}{\hbar}\frac{1}{\gamma - i(\omega-\epsilon_\lambda)}\Big[w_\lambda(t_{t_-}) - 2\frac{|d_{cv}|^2}{\hbar^2}\int_{-\infty}^{t_t}\mathrm{d}t'\ e^{[i(\epsilon_\lambda-\Omega)-\gamma](t_t-t')}\mathcal{E}_p^*(t') \times \int_0^\infty \mathrm{d}t\ e^{[i(\omega-\Omega)-\Gamma]t}\mathcal{E}_p(t+t_t) - 2\frac{|d_{cv}|^2}{\hbar^2}\int_0^\infty \mathrm{d}t\ e^{i(\omega-\Omega)t}\mathcal{E}_p(t+t_t) \times \int_0^t \mathrm{d}t'\ e^{-\Gamma(t-t')}e^{-[i(\epsilon_\lambda-\Omega)+\gamma]t'}\mathcal{E}_p^*(t'+t_t)\Big] \quad (31)$$

Excitonic Optical Stark Effect
Dynamic results



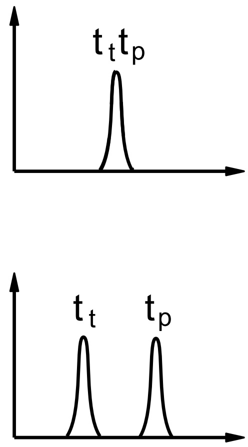


Figure: Differential absorption spectrum with probe pulse before pump pulse. Starting from bottom with 500 fs delay in 100 fs steps [Haug].

Excitonic Optical Stark Effect
Dynamic results

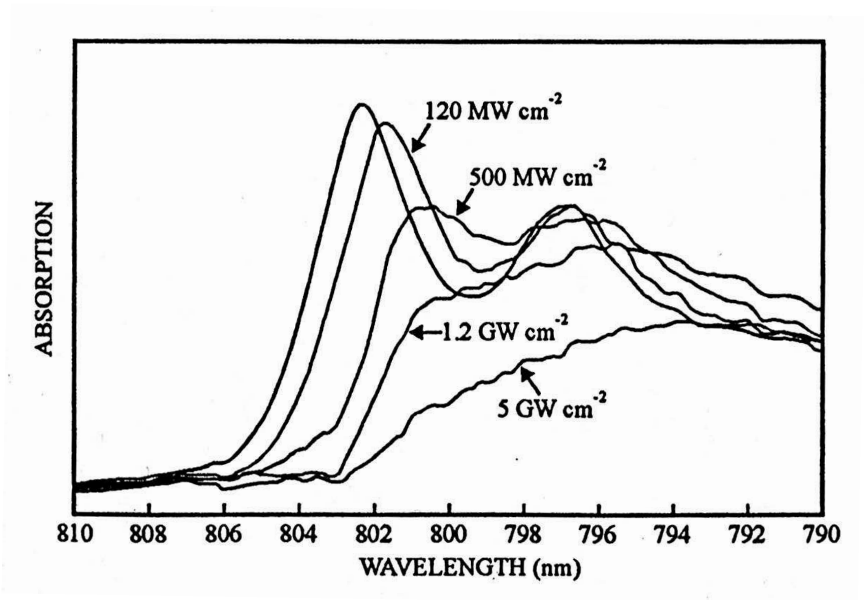


Figure: Experimental Absorption using 400 fs pulses for different pump pulse intensities[Schaefer].

References

[Ell] C. Ell, J. F. Müller, K. El Sayed, L. Banyai, and H. Haug.
Evaluation of the hartree-fock theory of the excitonic optical stark effect.
physica status solidi (b), 150(2):393–399, 1988.

[Haug] Hartmut Haug and Stephan W. Koch.
Quantum Theory of the Optical and Electronic Properties of Semiconductors.
World Scientific, London, 4. edition edition, 2004.

[Schaefer] Wilfried Schäfer and Marin Wegener.
Semiconductor optics and transport phenomena.
Springer, Berlin; Heidelberg, 2002.