Four Wave Mixing Studies of Polaron Dephasing in InAs/GaAs Self-Assembled Quantum Dots

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Outline

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- Introduction
  - Strong coupling and implications for carrier relaxation
- Polaron dephasing
  - Motivation, technique used
- Results
  - Experimental data
  - Calculation of acoustic phonon sidebands
  - Calculation of four-wave mixing signal: virtual and real transitions
  - Comparison of the theory and experiment
- Summary

InAs/GaAs QDs: Intraband spectroscopy

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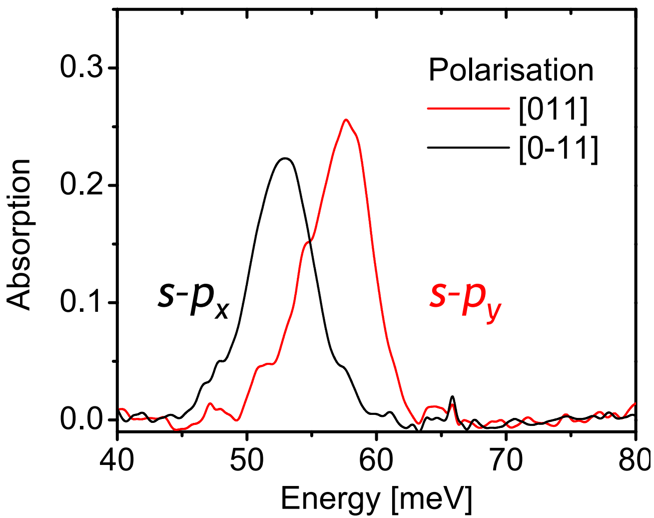

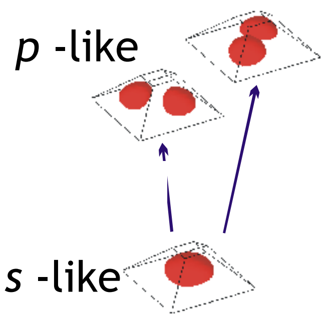

Intraband absorption

- Normal incidence geometry - in plane dipole
- Inhomogeneous broadening: FWHM ~5 meV
- Polarisation dependence due to QD wf anisotropy: splitting of *p*-states ~5 meV

Studies have shown that electrons and phonons in QDs are in the strong coupling regime – forming **polarons**

See for example:

S. Hameau *et al.*, Phys. Rev. Lett. **83** 4152 (1999)

X.-Q. Li & Y. Arakawa, Phys. Rev. B **57** 12285 (1998)

Implications for carrier relaxation

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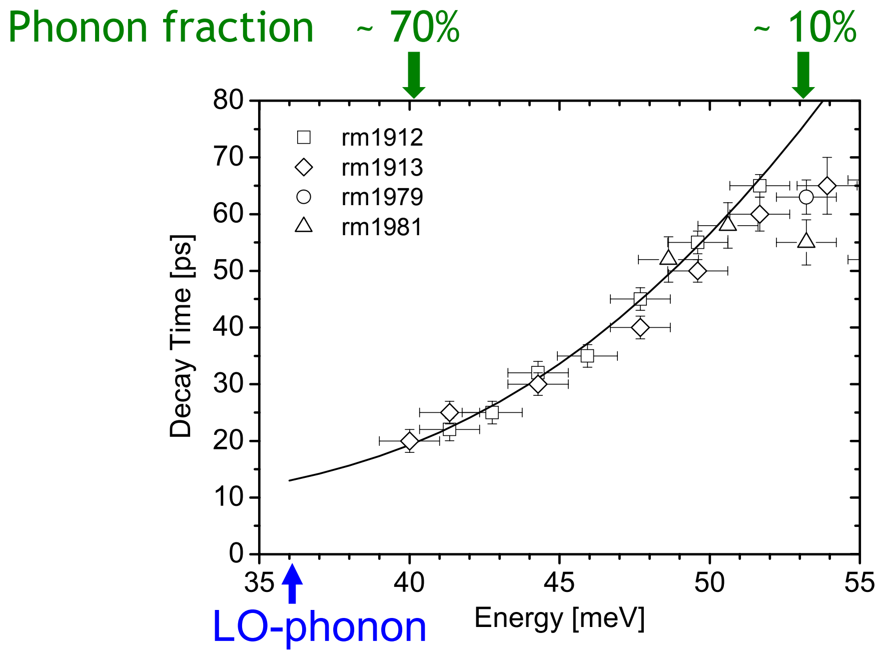
- Weak coupling picture does not apply, hence 'phonon bottleneck' does not exist

- Excited state polarons decay due to finite lifetime of the LO phonon - as the phonon fraction of the polaron increases the polaron lifetime decreases



X.-Q. Li & Y. Arakawa, Phys. Rev. B **57** 12285 (1998),

S. Sauvage *et al.*, Phys. Rev. Lett. **88** 177402 (2002),

E.A. Zibik *et al.*, Phys. Rev. B **70** 161305(R) (2004)

Polaron dephasing - motivation

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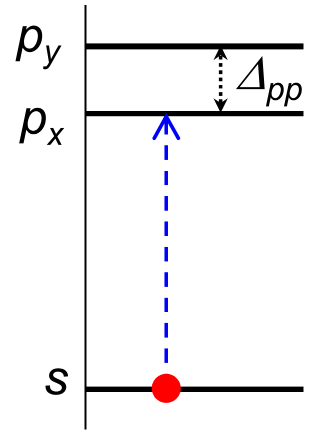
- Understanding dephasing processes due to the electron-phonon interaction in QDs is a key issue for many potential applications
- Provides homogeneous linewidth information - unlike interband case, intraband absorption measurements of single QDs not possible
- No holes present in the samples
- Simple 3-level energy structure makes easier to analyse the results and allows a clear picture of dephasing mechanisms to be obtained



Four Wave Mixing (FWM): Photon Echo

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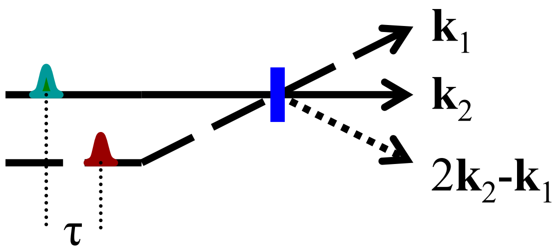
- Highly sensitive technique for studying the coherent behaviour of carriers in quantum dot systems



- Incident pulses $\mathbf{k}_1$ and $\mathbf{k}_2$; Delay time τ
- The photon echo ($2\mathbf{k}_2-\mathbf{k}_1$) is measured as a function of the time delay between the incident pulses and the dephasing time is deduced from the time decay of the coherent polarisation.
- Pulse duration ~ 1 ps

FWM comparison with Pump-Probe

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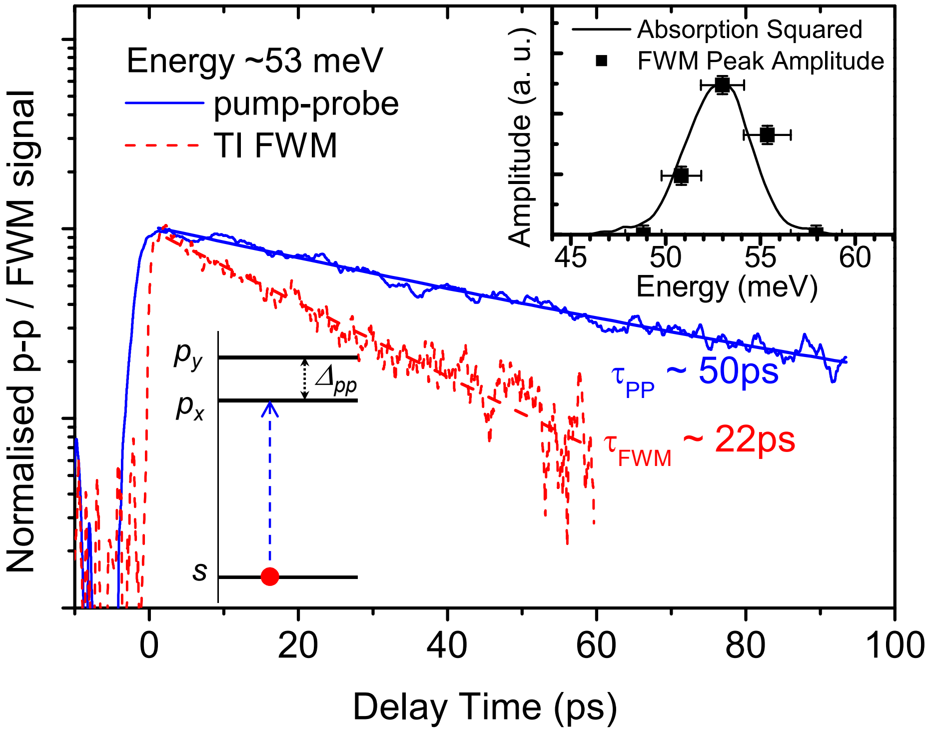
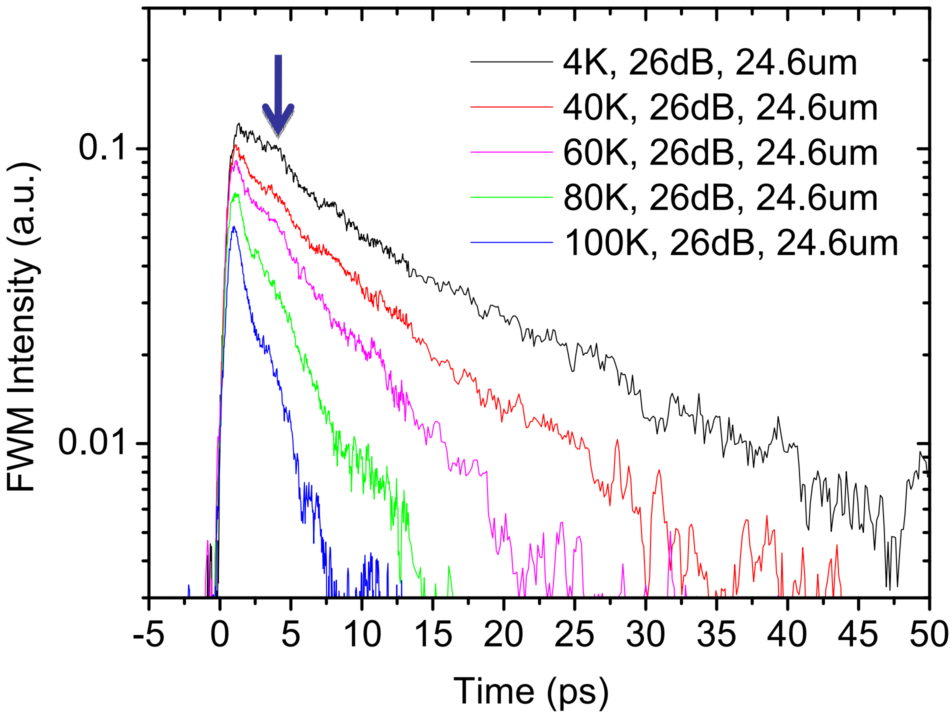


- Measurements performed in the χ^3 regime: ~250 pJ per pulse
- FWM amplitude follows dot linear absorption
- Very different decay times for pump-probe (~50ps) and FWM (~20ps)
- Inhomogeneous broadening: T_2=4xτ_{FWM}=88ps, close to $2T_1$ = 100 ps

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}$$

- Pure dephasing time: ~500ps, hence at low temperature the dephasing is mainly determined by *population relaxation*

Polaron dephasing results



- Polaron **dephasing time** (T_2) decreases dramatically with increasing temperature: from ~90ps at 10K to ~15ps at 100K (*polaron lifetime* T_1 decreases from ~50ps to ~40ps)
- This corresponds to decrease of the pure dephasing time from 500ps to ~18ps over the same temperature range

Theoretical approach

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- Polaron interaction with longitudinal acoustic phonons (deformation potential coupling) treated using the independent Boson model

- Deformation potential coupling matrix element

$$M_{\boldsymbol{q}}^{ij} = D_c \sqrt{\frac{\hbar q}{2\rho c_s V}} \langle i | e^{i\boldsymbol{q}.\boldsymbol{r}} | j \rangle$$

where $i = s, p_x, p_y$

- Absorption lineshape is given by

$$A(\omega) = Z e^{f(\omega)} = Z \left\{ \delta(\omega) + f(\omega) + \frac{1}{2}[f \otimes f](\omega) + \ldots + \frac{1}{p!}[f \otimes^{p-1} f](\omega) \right\}$$

Zero-phonon line *1*-phonon absorption/emission

p-phonon processes

where $f(\omega) = \sum_{\boldsymbol{q}} \frac{|M_{\boldsymbol{q}}^{p_x p_x} - M_{\boldsymbol{q}}^{ss}|^2}{\omega_q^2} [N(\omega) + \Theta(\omega)]\, \delta(\omega - \omega_q)$

and the weight of the zero phonon line $Z = exp\left[- \int_{-\infty}^{+\infty} \mathrm{d}\omega f(\omega) \right]$

Acoustic phonon sidebands

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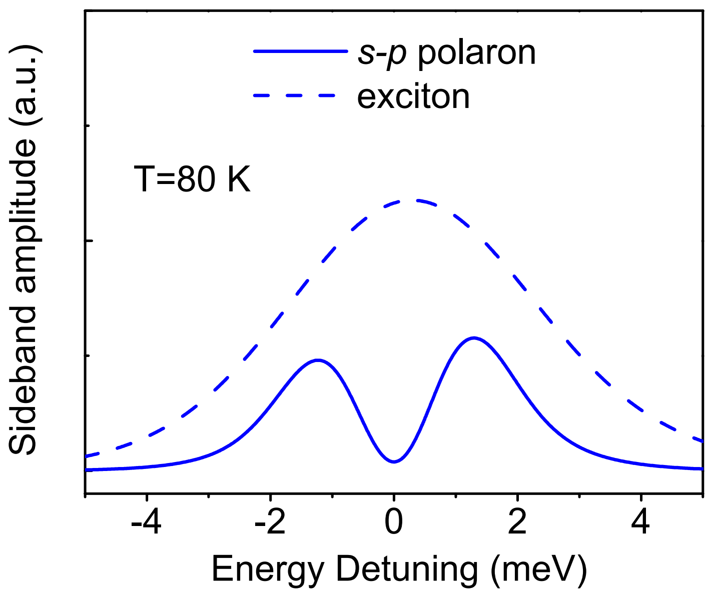
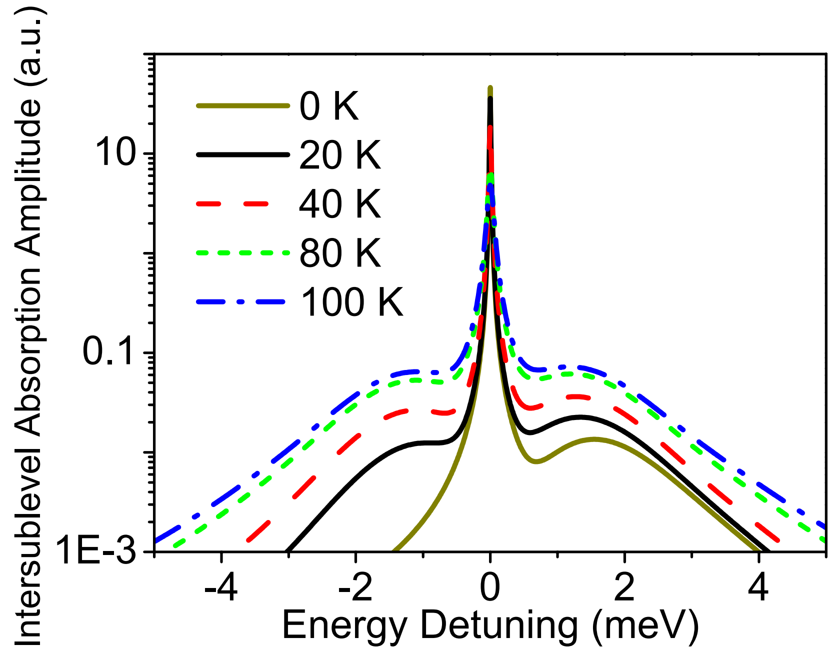
Interband: $$f(\omega)=\sum_{\boldsymbol{q}}\frac{|M_{\boldsymbol{q}}^{s_e s_e}+M_{\boldsymbol{q}}^{s_h s_h}|^2}{\omega_q^2}\left[N(\omega)+\Theta(\omega)\right]\delta(\omega-\omega_q)$$

$$M_{\boldsymbol{q}}^{s_e s_e}+M_{\boldsymbol{q}}^{s_h s_h}=\sqrt{\frac{\omega_q}{2c\rho}}\left(D_c\langle s_e|e^{i\boldsymbol{q}.\boldsymbol{r}_e}|s_e\rangle\ +D_v\langle s_h|e^{i\boldsymbol{q}.\boldsymbol{r}_e}|s_h\rangle\ \right)$$

-7.2 eV (for D_c) -1.3 eV (for D_v)





Four wave mixing – calculations

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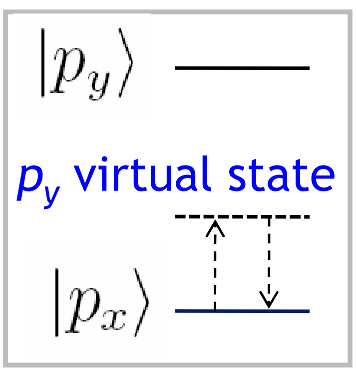
- FWM dynamics calculated for excitation in resonance with the s to p_x transition, taking into account polaron decay to the *s* state, real and virtual transitions to the p_y state, as well as the presence of phonon sidebands
- Broadening of the zero phonon line due to real and virtual phonon transitions from p_x to p_y

Absorption+Emission

$$\Gamma_2^* = \frac{1}{2\pi}\int_0^{+\infty} d\omega \frac{\Gamma_{pp}^2(\omega)N(\omega)\left[N(\omega)+1\right]}{(\omega-\Delta_{pp})^2+\left(\frac{\Gamma_{pp}(\omega)\left[N(\omega)+1\right]}{2}\right)^2}$$

$$+\frac{\Gamma_{pp}^2(\omega)N(\omega)\left[N(\omega)+1\right]}{(\omega+\Delta_{pp})^2+\left(\frac{\Gamma_{pp}(\omega)N(\omega)}{2}\right)^2}$$



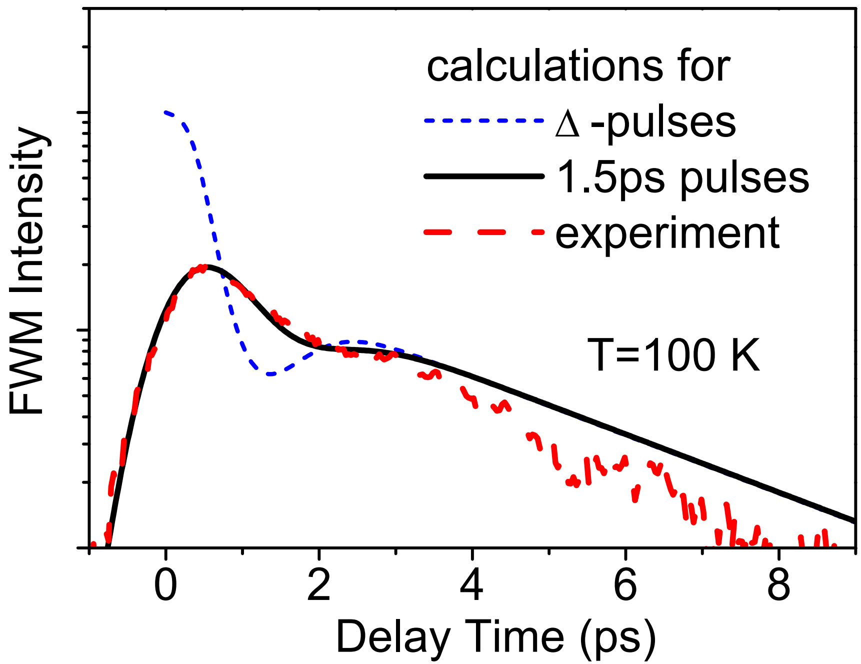
Emission+Absorption

where $\Gamma_{pp}(\varepsilon) = 2\pi\sum_{\boldsymbol{q}} |M_{\boldsymbol{q}}^{p_x p_y}|^2\delta(\varepsilon - \hbar\omega_q)$

Four wave mixing – calculations

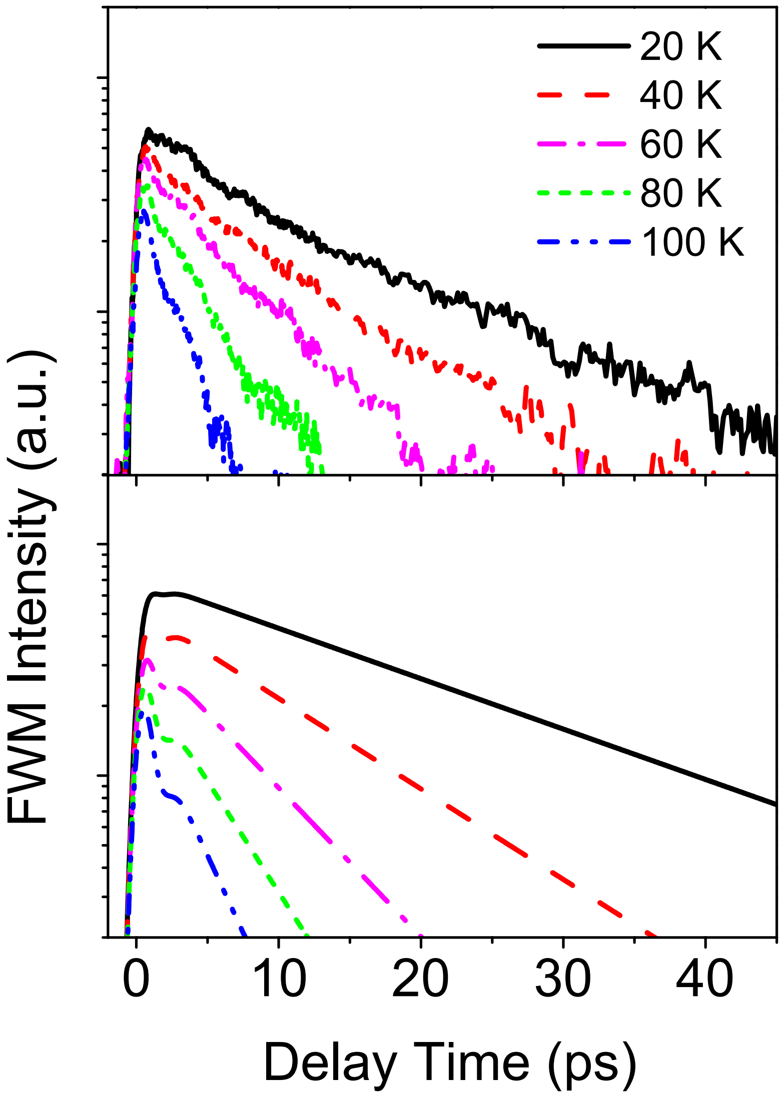
- Intensity of the FWM response to a delta pulse as a function of the delay time between the 2 pulses:

$$I(t) \propto \Theta(t) \exp\left[-2\Gamma_2 t - 16\int_{-\infty}^{+\infty} \mathrm{d}\varepsilon f(\varepsilon) \sin^2\left(\frac{\varepsilon t}{2\hbar}\right)\right]$$



- Presence of acoustic sidebands results in oscillatory behaviour in the FWM signal shortly (< 5 ps) after resonant excitation of the lowest energy conduction band transition, which is due to coherent acoustic phonon generation

FWM – temperature dependence



- Excellent agreement found between our measured and calculated FWM dynamics

FWM – temperature dependence

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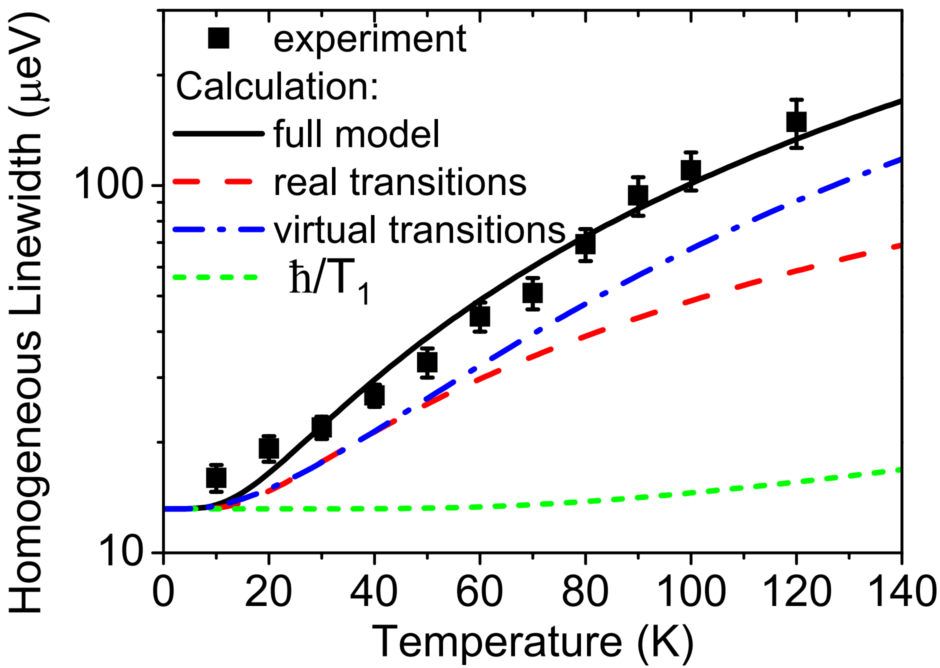

- Very good agreement between calculated total linewidth ($\Gamma_2 = \Gamma_1 + \Gamma_2^*$) of the zero phonon line and values deduced from experiment
- Virtual transitions dominate dephasing at high temperature due to the quadratic dependence on phonon occupation number

Summary

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- First studies of polaron dephasing processes in InAs dots using far infrared transient four wave mixing (FWM) spectroscopy
- Acoustic phonon sidebands ~1.5 meV apart from zero-phonon line (Γ_{hom}~20 μeV)
- Oscillatory behaviour in the FWM signal shortly (< 5 ps) after resonant excitation is due to coherent acoustic phonon generation
- Subsequent single exponential decay yields long intraband dephasing times of ~90 ps
- Both real and virtual acoustic phonon processes are necessary to explain the temperature dependence of the polarisation decay