Spin Coherence and Spin Memory Effects in InAs Self-Assembled Quantum Dots

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1. Linearly polarised eigenstates
2. Single dot spectroscopy
3. Coherent and incoherent spin effects, relatively robust dephasing and polarisation (spin) memory (ensembles)
4. Strong effect of annealing. Control of the fine structure splitting
5. Conclusions

- Self assembled quantum dots important for both physics and applications
- Quasi-0D systems in the solid state. ‘Atom-like’
- Strong confinement <u>and</u> high radiative efficiency
- Embedded in semiconductor matrix. Wide variety of semiconductor devices, processing technology
- Long spin lifetimes, dephasing times due to discrete density of states (quantum information applications)?

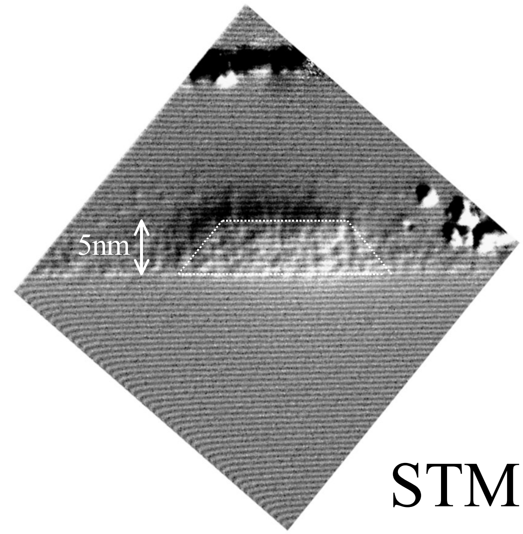
Acknowledgements

Ultrafast: AI Tartakovskii, J Cahill, MN Makhonin, AM Fox, JPR Wells

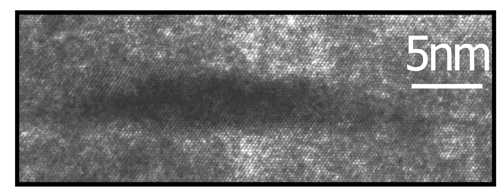
Continuous wave: JJ Finley

Growth: MJ Steer, H Liu, M Hopkinson

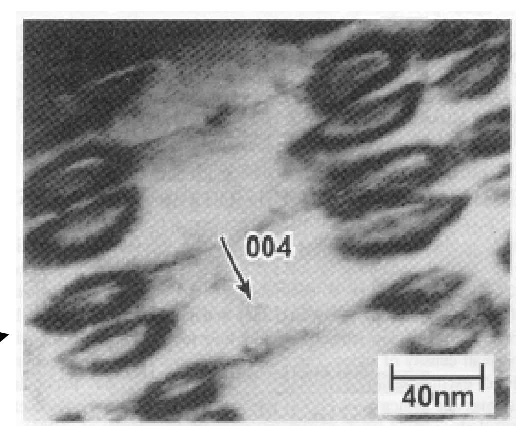
Theory: D M Whittaker



STM



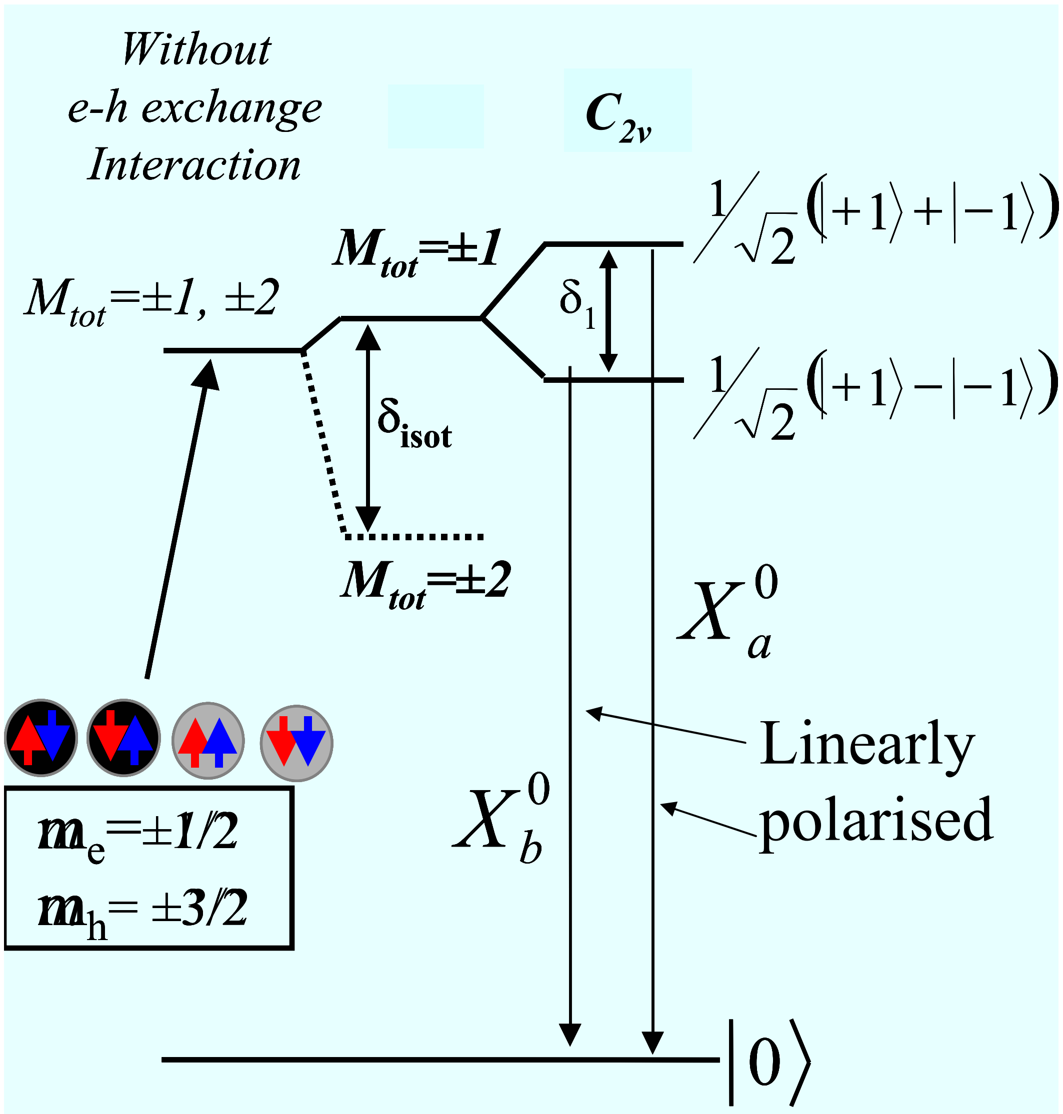
TEM



Goldstein APL 47, 1099, 1985

Spin Phenomena, cw (single dots) and ultrafast (ensembles)

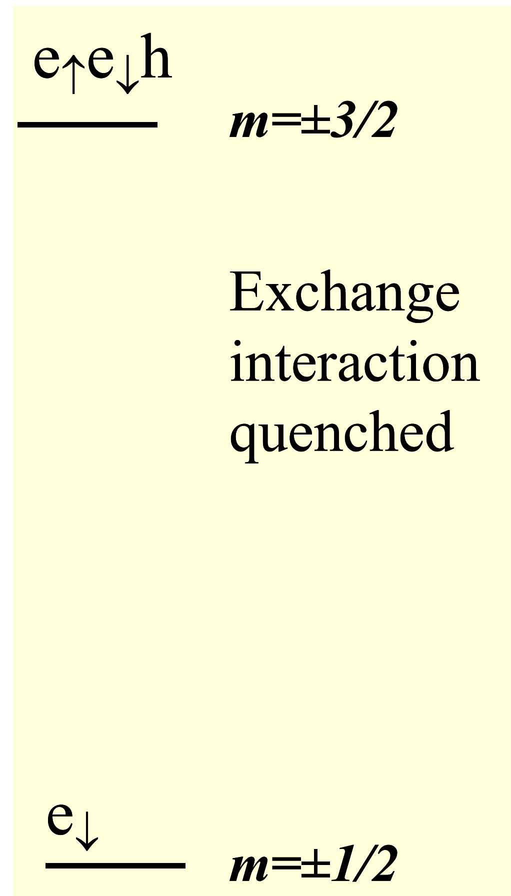
Neutral exciton X° e, h



(a)

Negatively charged exciton

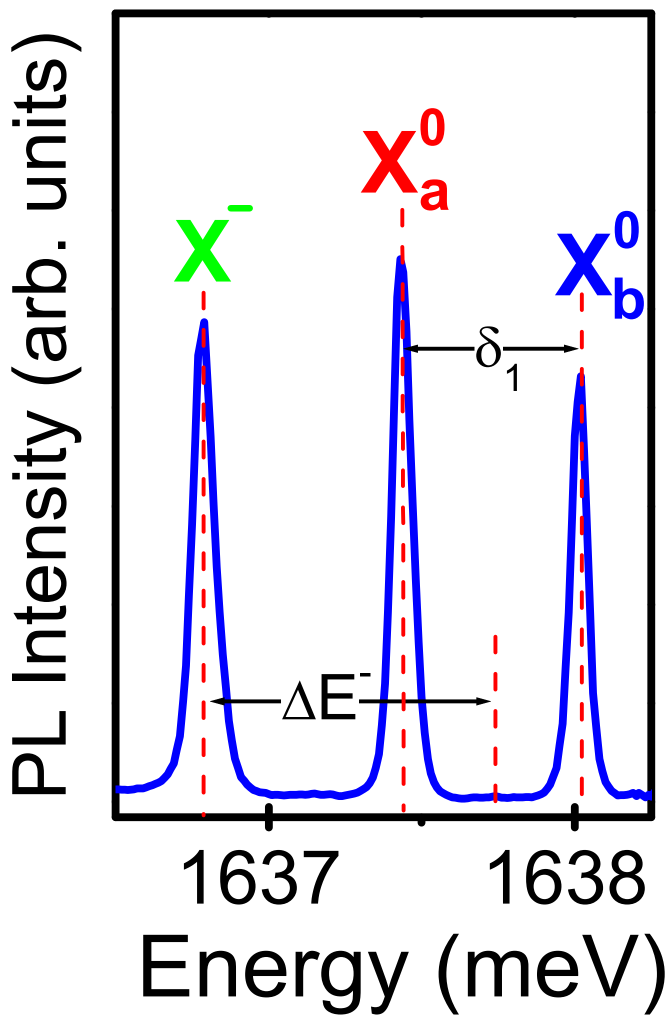
X⁻ 2e, h



(b)

InAs-$Al_{0.6}Ga_{0.4}As$ QDs

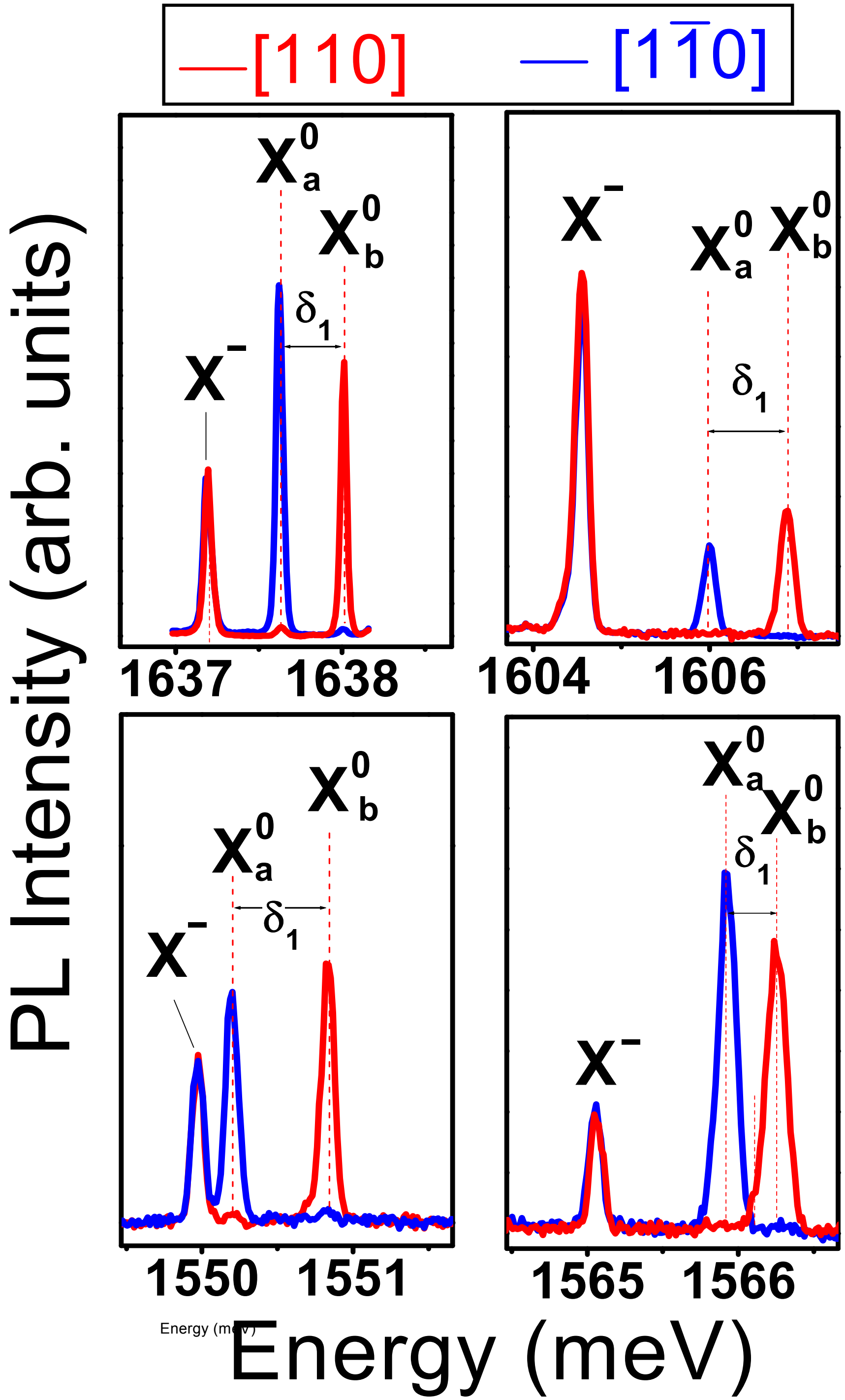
Finley et al PRB 66, 153316, 2002



- Red emitting, Al-containing dots
- **Very large (anisotropic) exchange splittings, δ_1~0.8meV**

Kulakovskii et al, PRL 82, 1780, 1999, Bayer et al, PRL, 82, 1748, 1999, Bayer et al PRB 2002

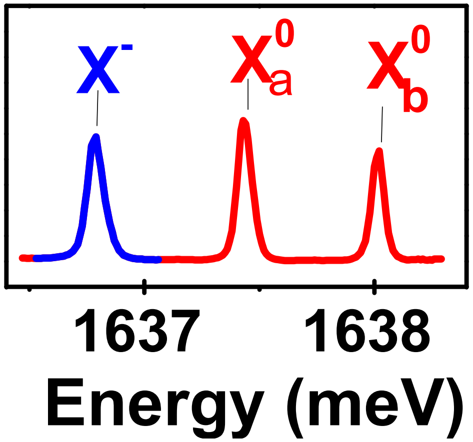
And early papers on bound excitons in bulk semiconductors (*e.g. Morgan and Morgan Phys Rev B1, 739, 1970)*



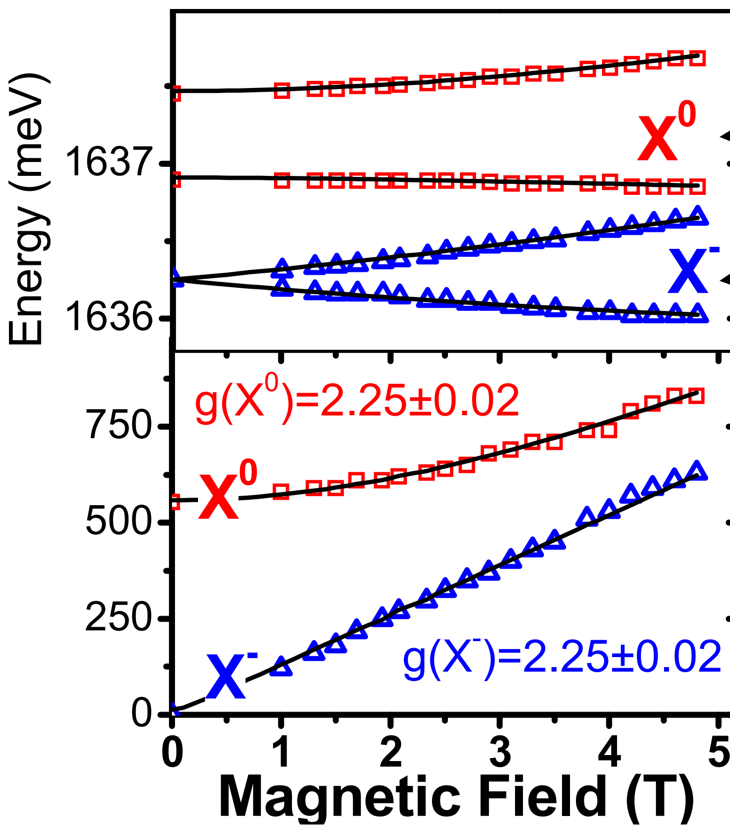
4 different dots

Strong splitting and linear polarisation observed for neutral exciton X^0

X^- unsplit, unpolarised



Zeeman Splitting



$X^0_{a,b}$ doublet

- Good fits to **peak positions**, **polarisation** and **splitting** from diagonalisation of HH-spin Hamiltonian in bright exciton basis
- Identifies $X^0_{a,b}$ as bright exciton states in dot with <110> axial

$$\Delta E(B) = \sqrt{\delta_1^2 + \left(g_{ex}^{X^o} \mu_B B\right)^2}$$

$$\Delta E(B) = g_{ex}^{X^o} \mu_B B$$

X⁻ g-factor **identical** to X^0 doublet to high experimental accuracy ($\pm$1%)

Observations consistent with X^- being charged exciton...

Ultrafast pump-probe

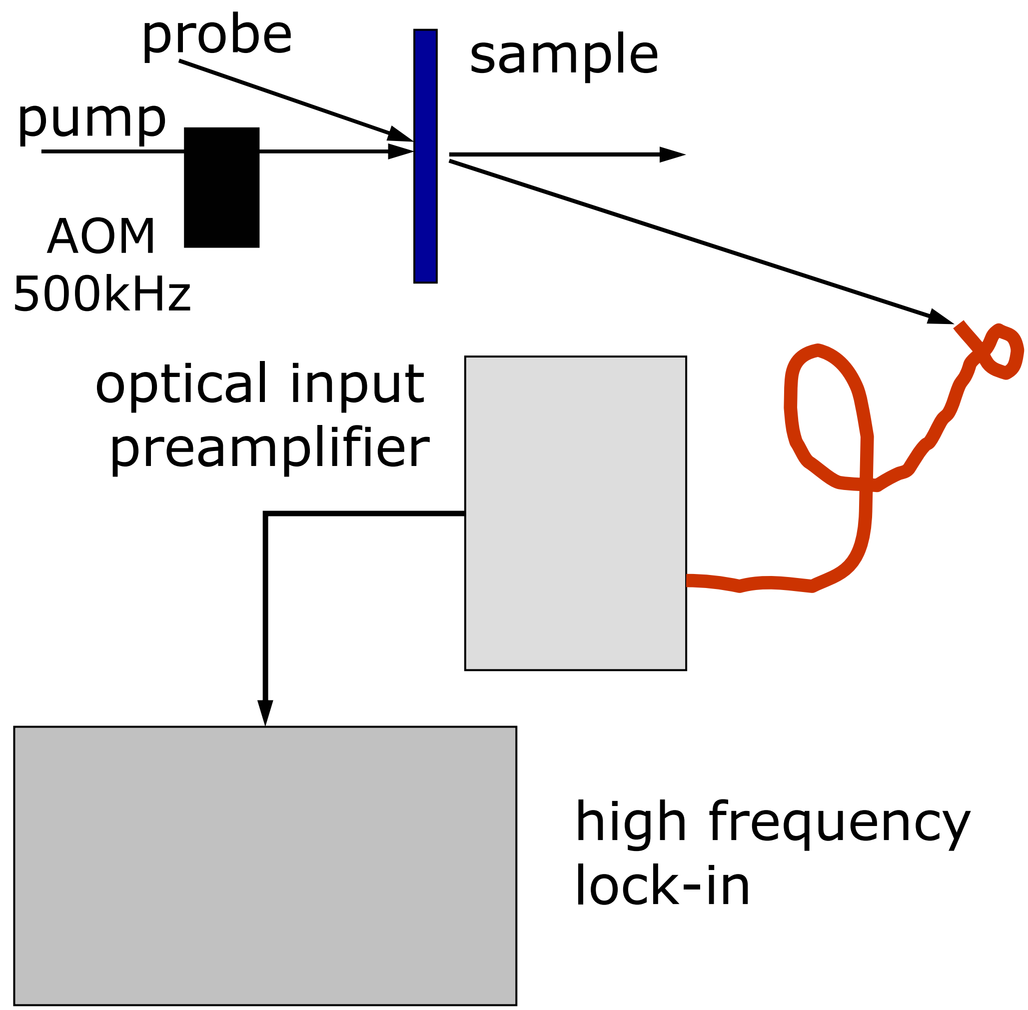
Coherent and incoherent effects

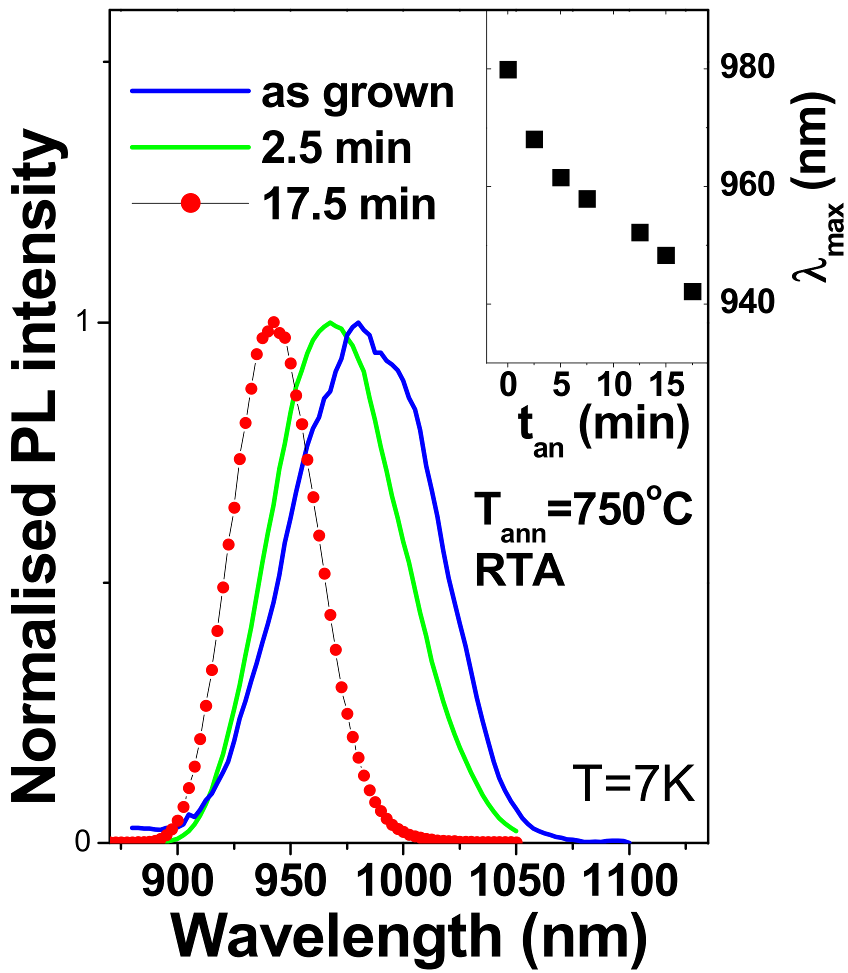
3 - 16 layer dot ensembles

AI Tartakovskii et al, PRL 2004, in press

Pump-probe experiment

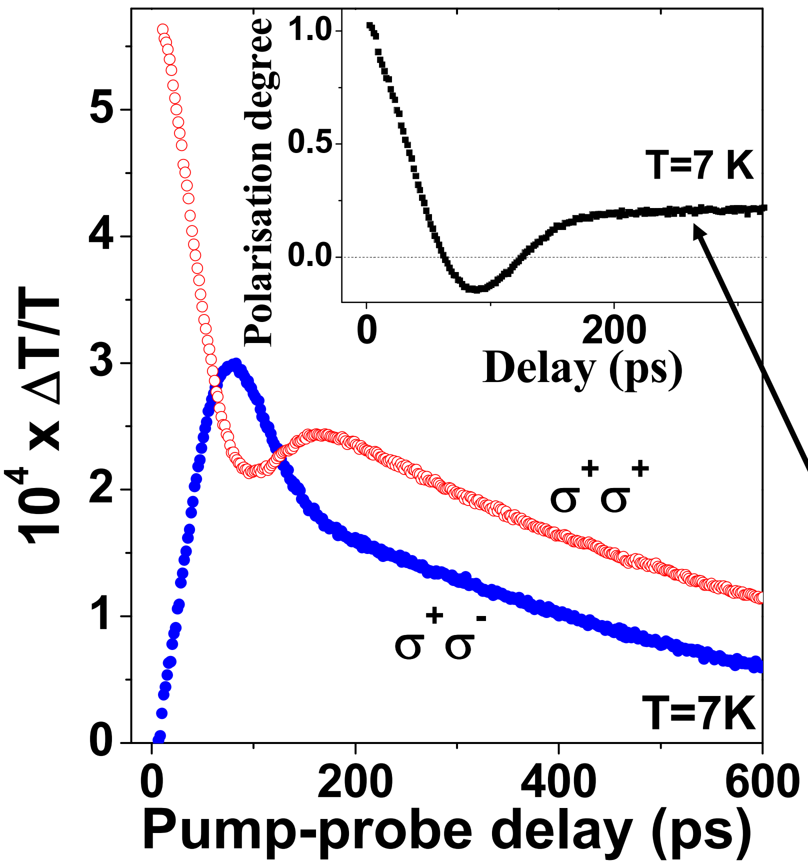
Near normal incidence





Degenerate pump and probe at ~980nm

Spin Coherence: Pump-Probe Measurements on Quantum Dot Ensembles



16 layers, uncoupled

Also see Paillard et al, PRL 86, 1634, 2001

σ^+ pump, co and cross circularly polarised probe

Two regimes:

1. Oscillations up to ~200psec
2. Exponential decay up to several nsec

Quantum beats: arise from ground state splitting

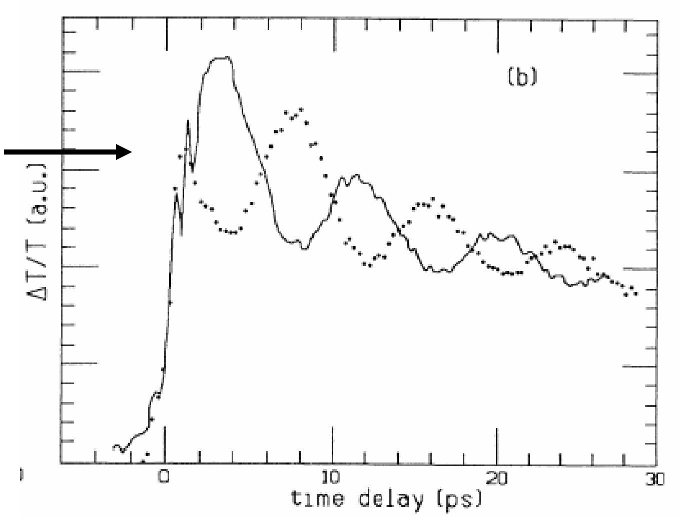
'High' temperature, several nanosecond, polarisation memory based on charged dots (incoherent). Increases slightly with time!

$$\psi_{lin}^{+,-} = 1/\sqrt{2}\left(|1\rangle \pm |-1\rangle\right)$$

Circularly polarised pump excites coherent superposition of $|1\rangle \pm |-1\rangle$. Eigenstates decay at slightly different rates, and give rise to beats.

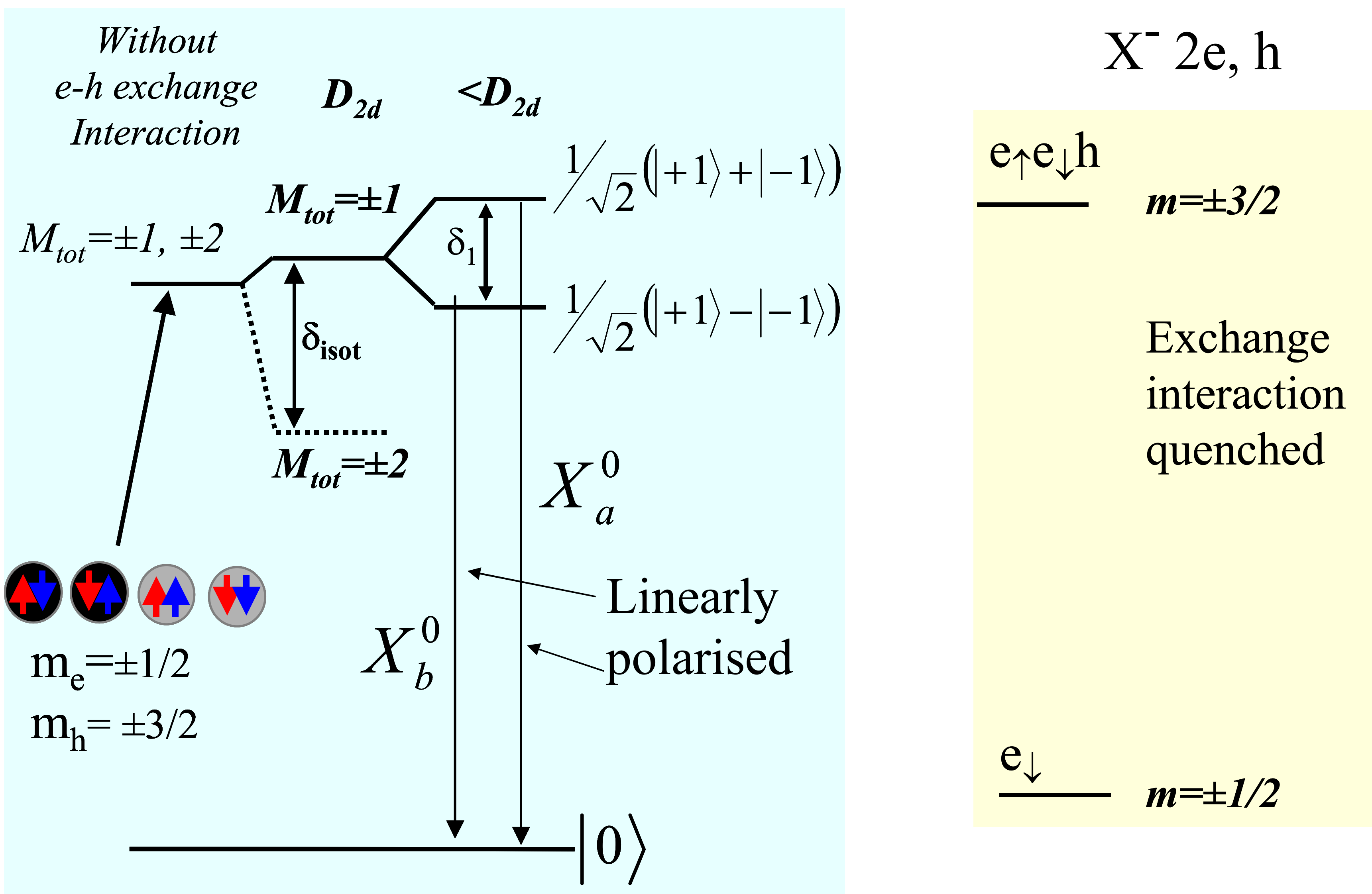
Quantum beats observed at frequency given by splitting δ_1 = 18μeV, period ~140psec. Reasonably typical for InAs dots

Four wave mixing in a co-linear geometry (see e.g. Bar-Ad and Bar-Joseph, PRL 66, 2491, 1991 for QWs in B-field, and e.g. Lenihan, Steel et al PRL 88, 223601, 2002 for dots)

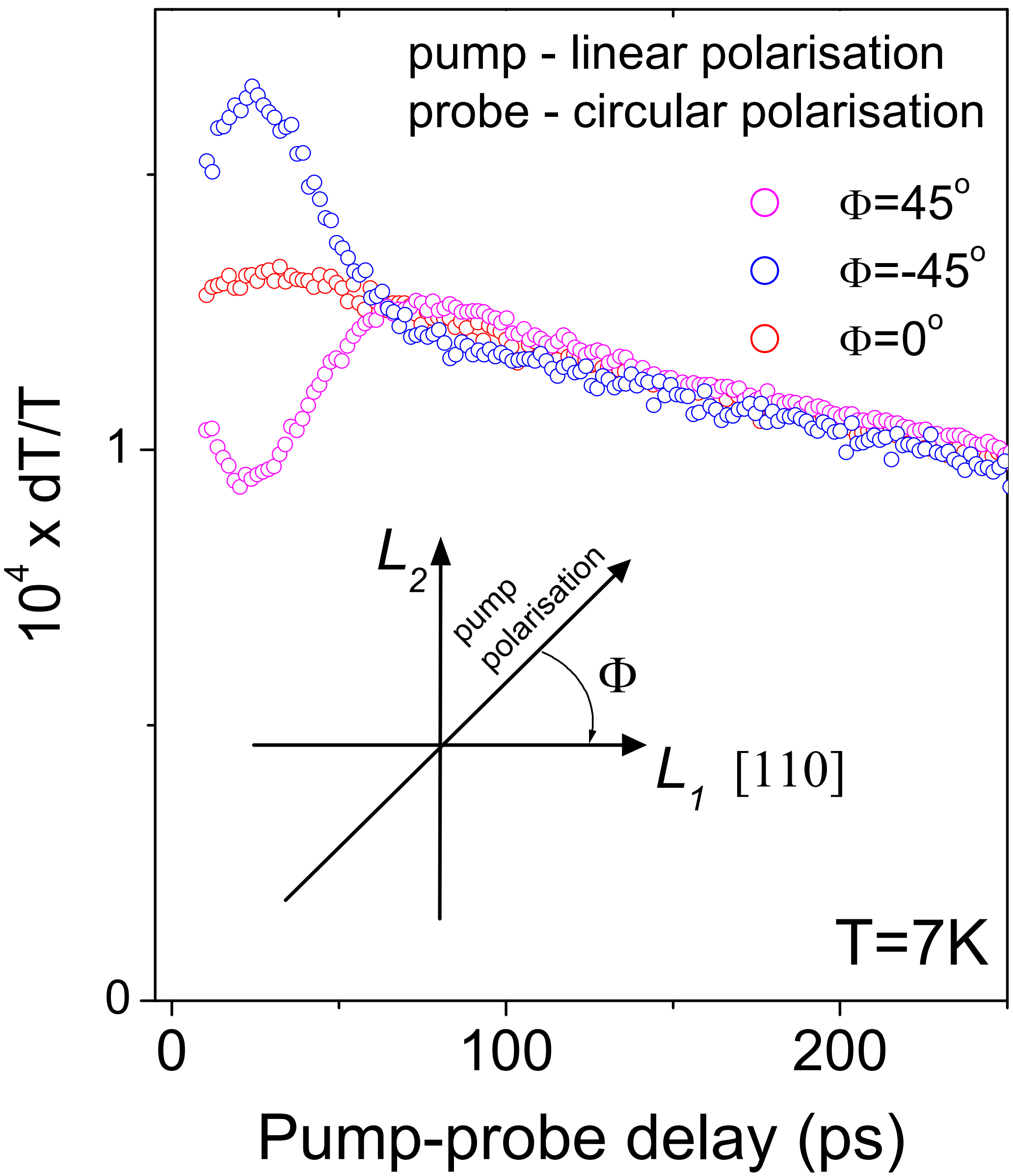


Neutral exciton X^o e, h

Negatively charged exciton X^- 2e, h



(a) (b)



Beats absent for excitation parallel to either of (110) directions – since only one of linearly polarised eigenstates then excited – thus no beating

Supports splitting as due to anisotropic exchange

Time Dependence of Pump-Probe Signal

coherent term

$$I_{\sigma^+\sigma^+/\sigma^+\sigma^-} = e^{-t/\tau_X} \pm \overbrace{e^{-t/T_{coh}} e^{-\delta^2 t^2} \cos(\frac{E_{FS}}{\hbar}t)} + \frac{(1\pm 1)}{4} A_{ch} e^{-t/\tau_{chX}}$$

Population decay incoherent

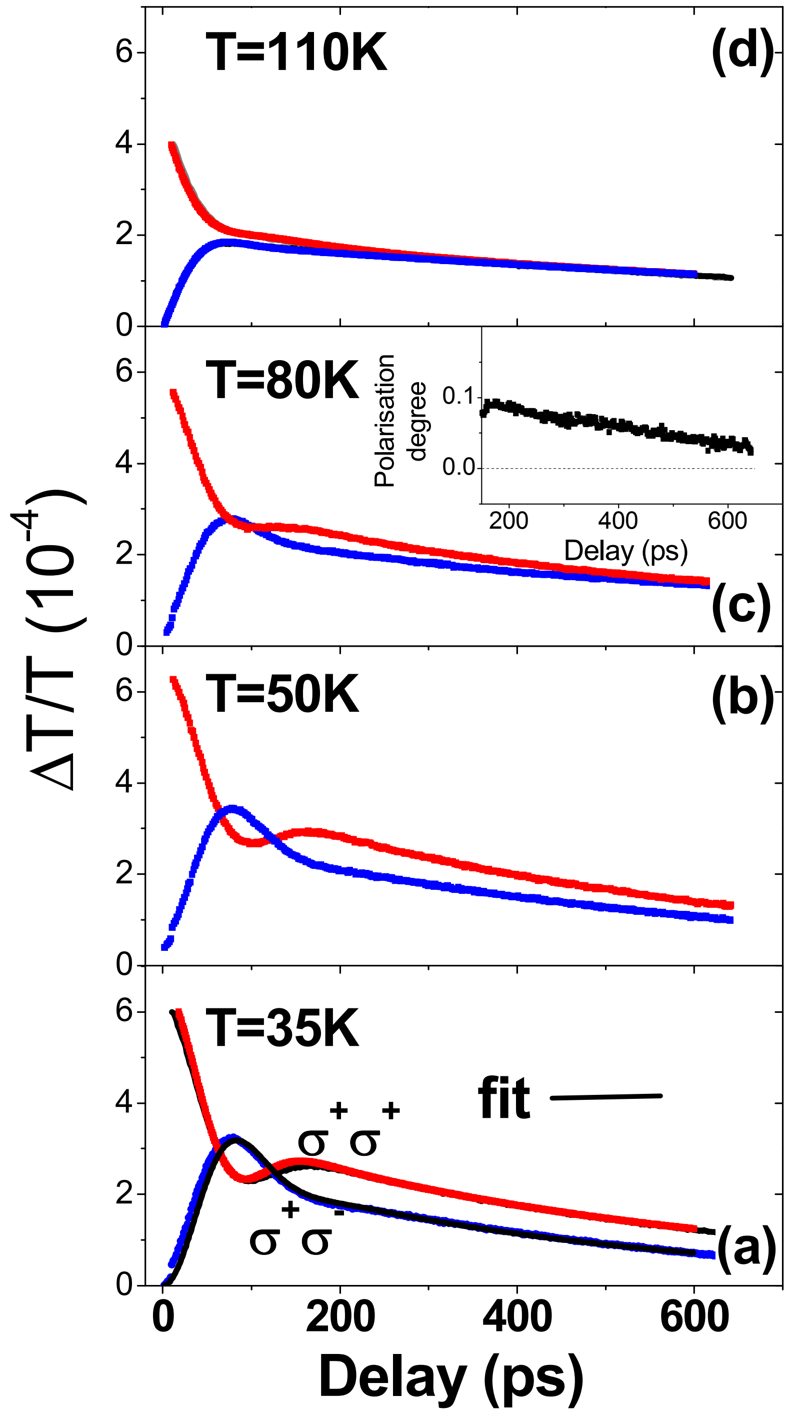
Dephasing

Inhomogeneous distribution, δ=6μeV

beats

Fraction of population which shows no beats and contributes to $I_{\sigma^+\sigma^+}$

M Mitsunaga and CL Tang, Phys Rev A35, 1720, 1987

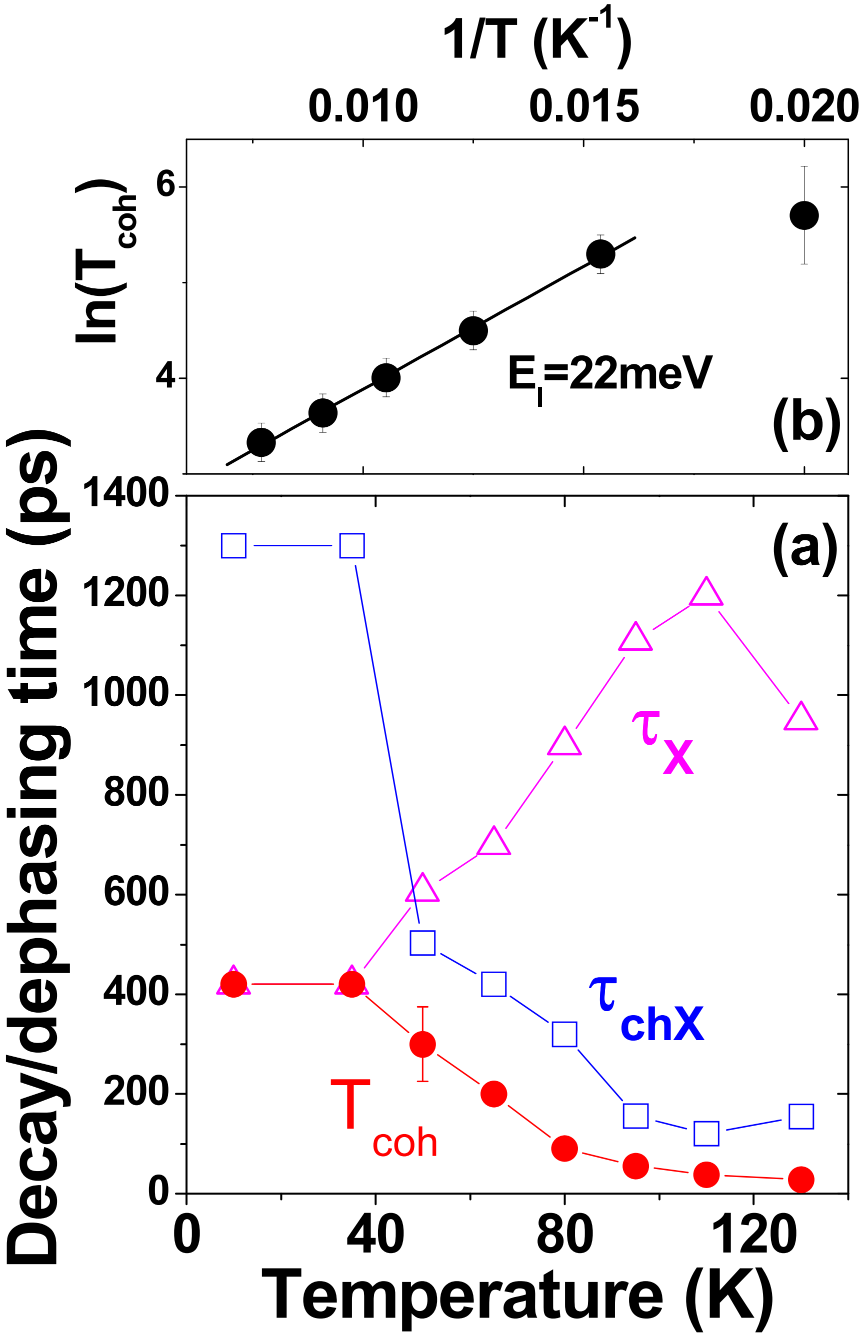


No significant temperature dependence of beats up to 55K

Inhomogeneous broadening dominates

Then beats are progressively damped

T_{coh} (T_2) at 130K ~5 larger than in FWM experiments (exciton decoherence, Borri et al)



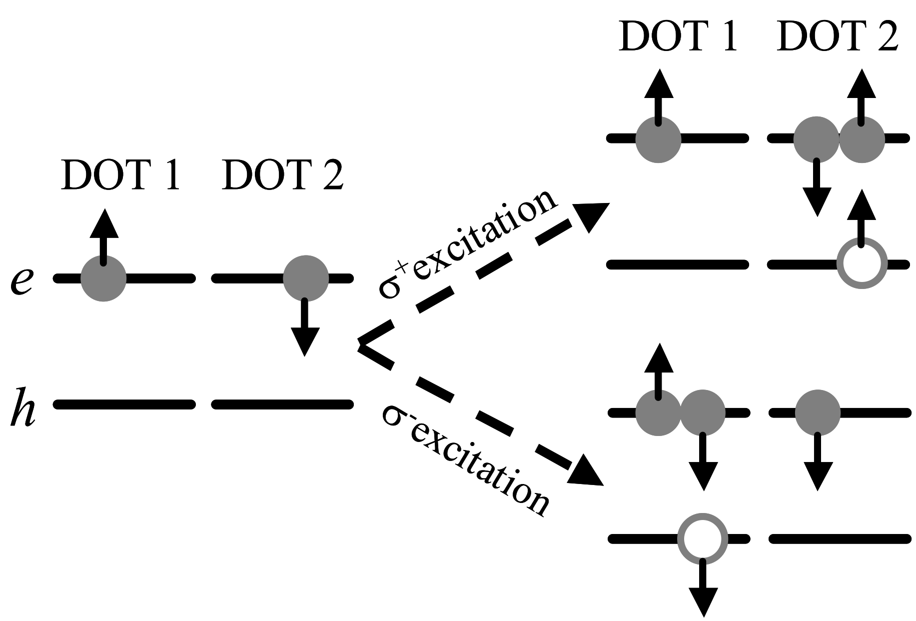
Spin dephasing time ~30psec at 130K

Factor 5-10 longer than exciton dephasing

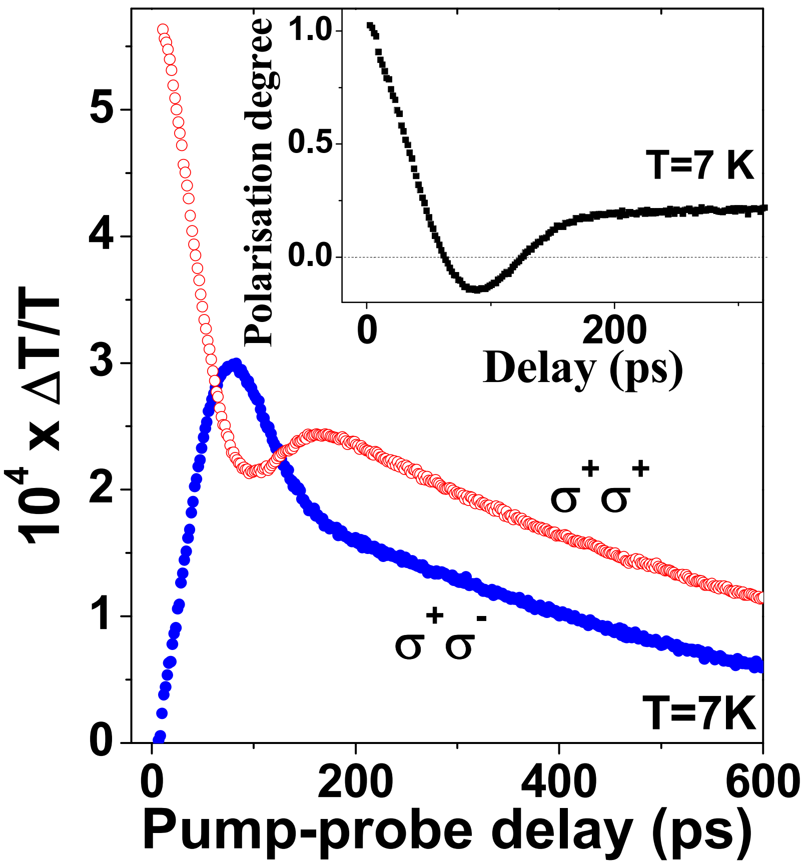
Activation energy ~22meV

Polarisation (Spin) Memory (difference in magnitude of σ^+, σ^- probe signals)

Can only fit pump-probe signals with an additional term arising from dots with no ground state spitting (no beats)



- For σ^+ excitation, only dot 2 can be excited
- Creates charged exciton, no exchange, no beats
- When probed with σ^+, absorption is blocked, and pump-probe signal
- σ^- probe unchanged by σ^+ pump absorption, hence no modulated pump-probe signal
- Hence means to probe charged dot fraction



10^4 x ΔT/T
Pump-probe delay (ps)
σ+σ+
σ+σ-
T=7K
Polarisation degree
Delay (ps)
T=7 K

coherent term

$$I_{\sigma^+\sigma^+/\sigma^+\sigma^-} = e^{-t/\tau_X} \pm e^{-t/T_{coh}} e^{-\delta^2 t^2} \cos(\frac{E_{FS}}{\hbar}t) + Ae^{-t/\tau_{chX}}$$

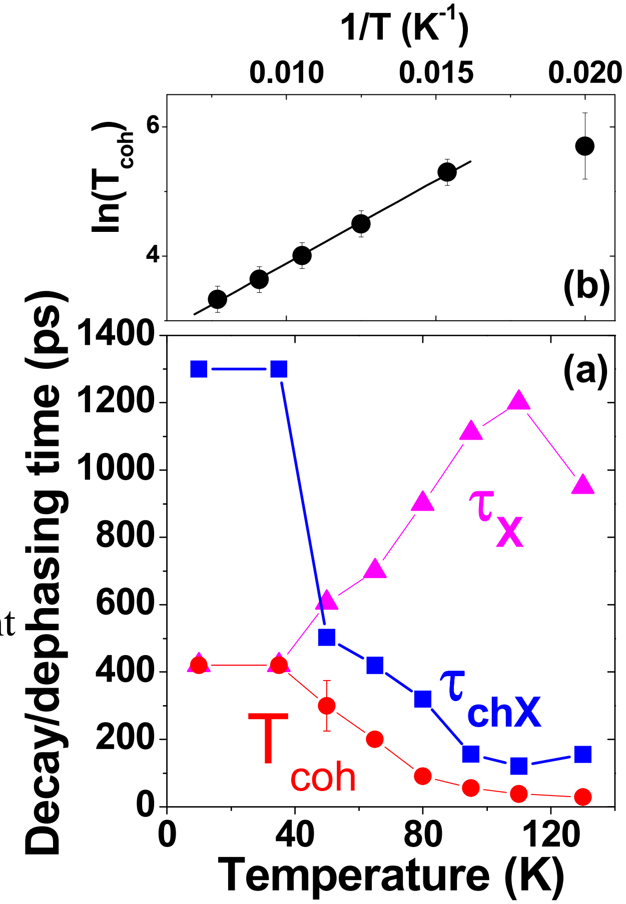
Population decay incoherent

Dephasing

Inhomogeneous distribution, δ=6μeV

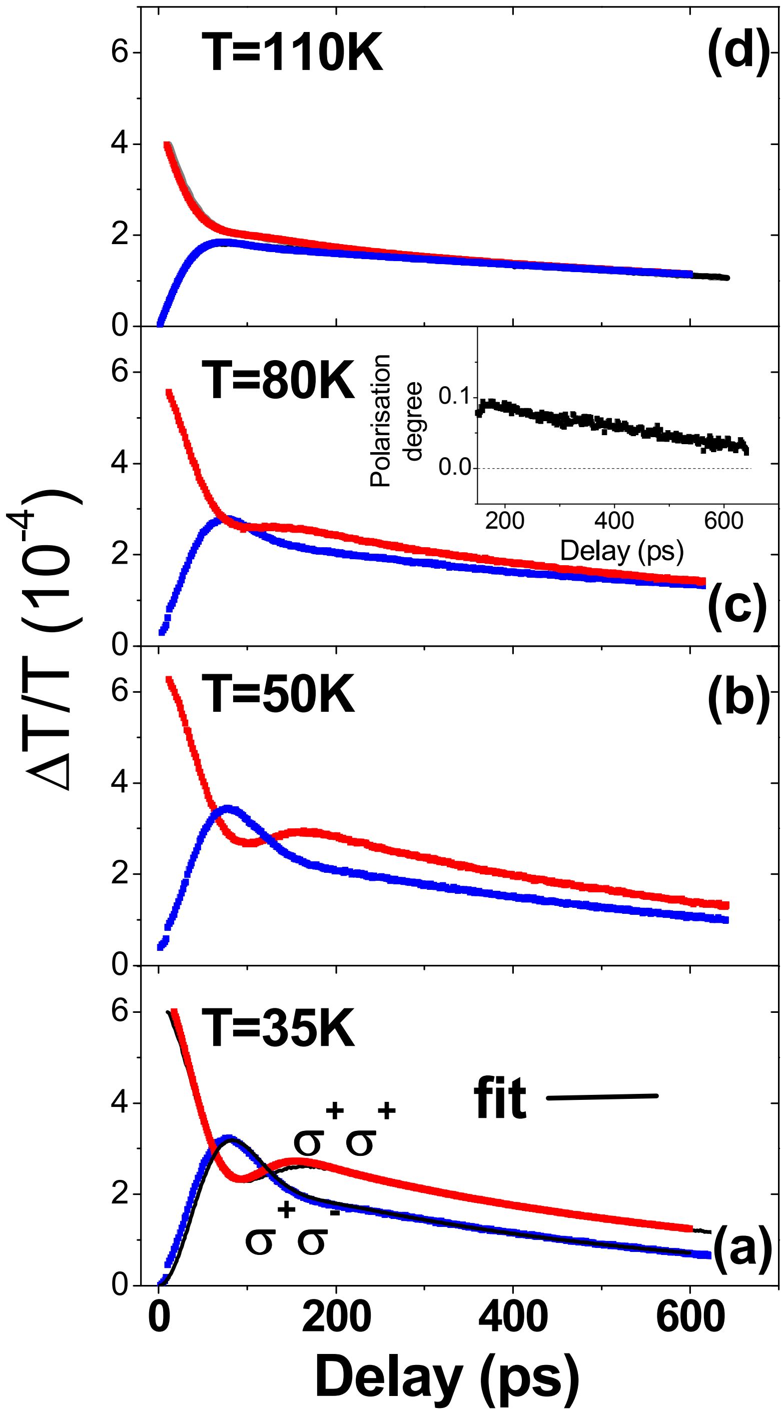
beats

Charged exciton decay, incoherent



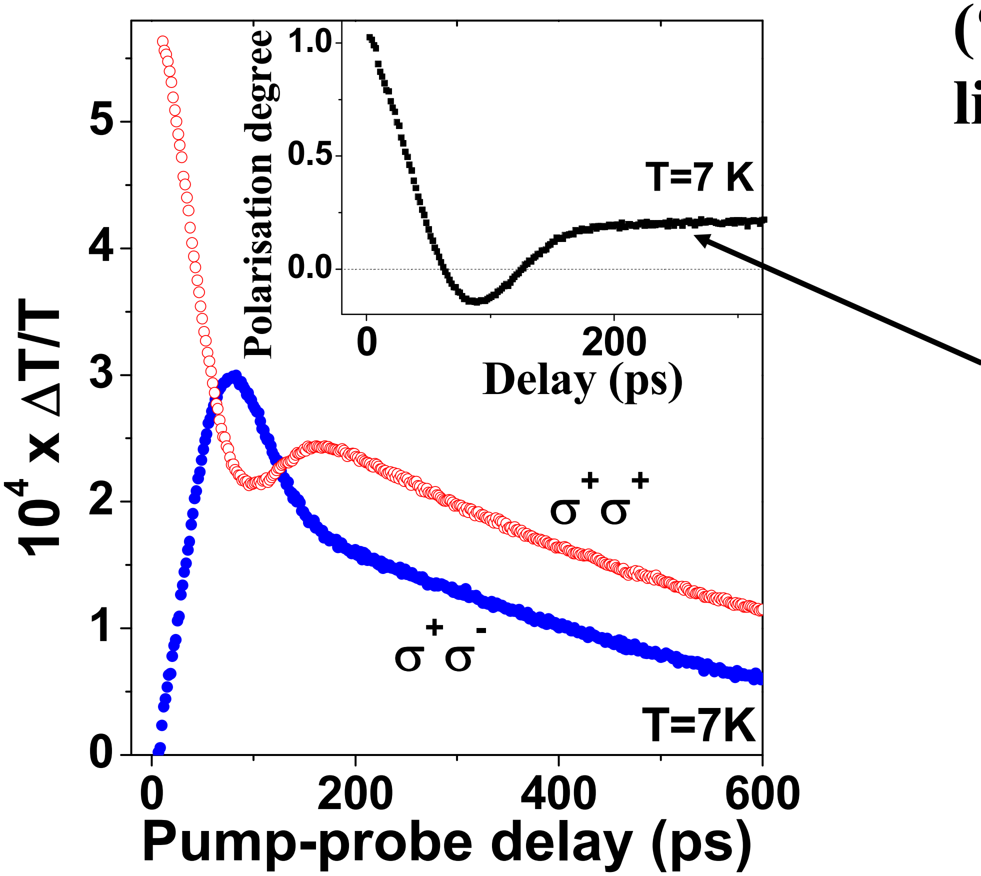
In addition to T_{coh} versus T, can also deduce temperature dependence of exciton and charged exciton lifetimes

$\tau_X > \tau_{chX}$ for T<50K. Leads to polarisation (spin) memory



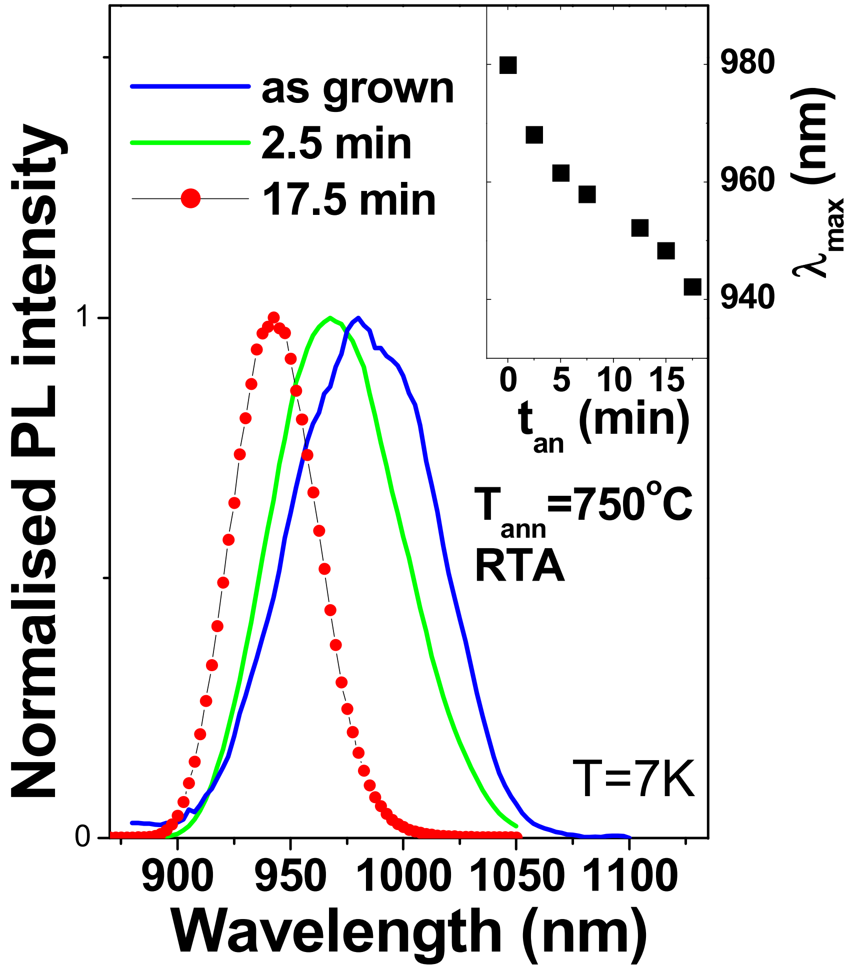
- Polarisation memory quenched with increasing T. Visible up to 100K.
- For T>50K, $\tau_X < \tau_{chX}$
- Notable that all three time constants show similar onset of temperature dependence (E_I ~22meV from T_{coh}).
- Excitation to hole excited state?
- Increase stability by using deeper dots

Degree of polarisation ('spin memory') very long lived >10nsec



Note that polarisation degree increases slightly with time at low temperature since $\tau_X < \tau_{chX}$

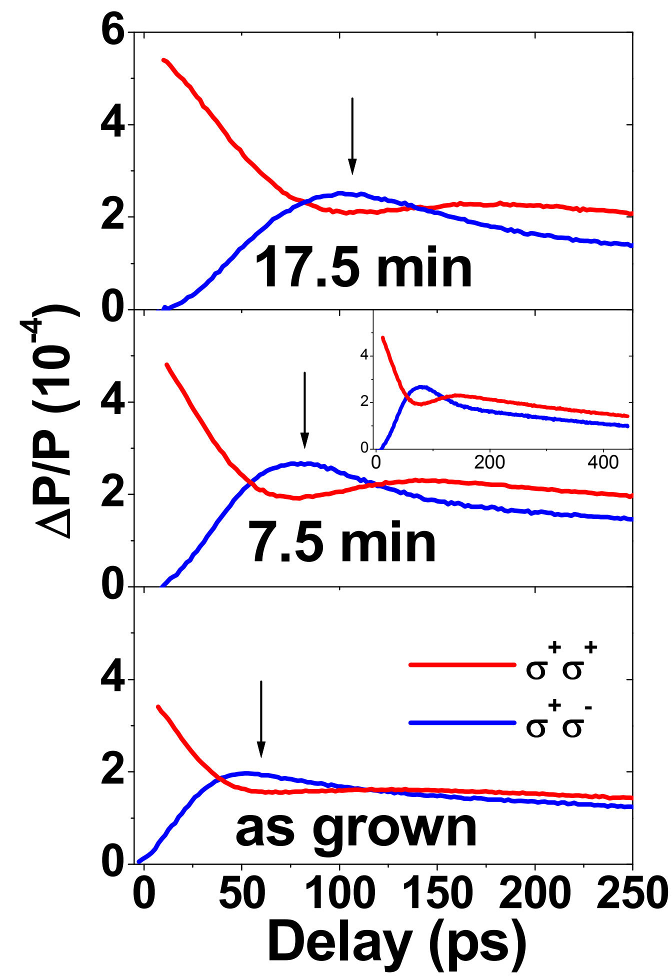
Effect of Annealing

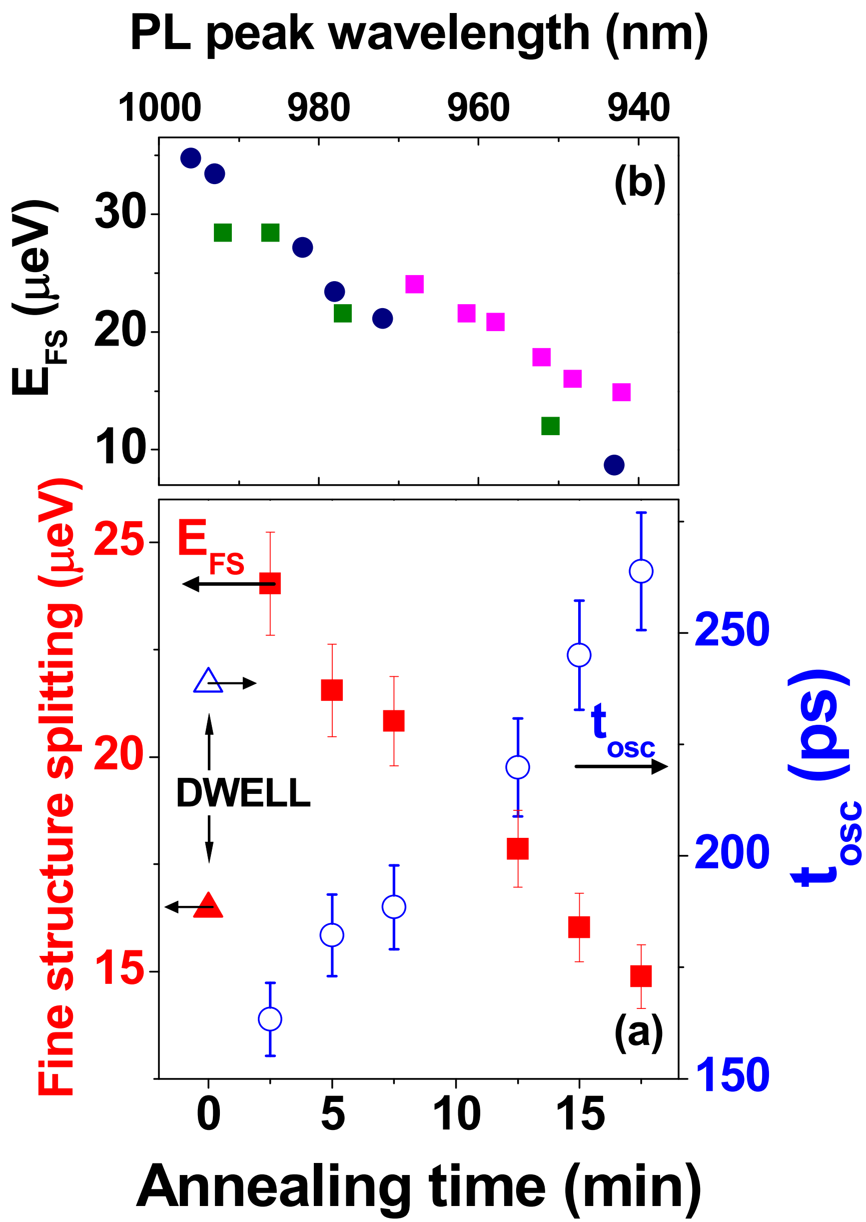


Shift to higher energy with annealing: Murray et al, APL 71, 1987, 1997, Leon et al 69, 1888, 1996. Intermixing, as for QWs, lowers barrier height, effective size increase

Two effects:

1. Beat period increases, fine structure splitting decreases





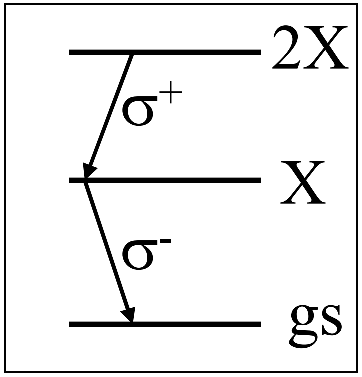
Ground state splitting decreased from 35 to 8μeV for only 17.5mins annealing

Langbein et al (*PR B69, 161301, 2004*) have observed reduction to 6 μeV

Possible means to create polarisation entangled photon source (from biexciton, exciton decay), Santori et al PR B66, 043508, 2002

At moment prevented by asymmetry splitting

Need splitting less than homogeneous linewidth



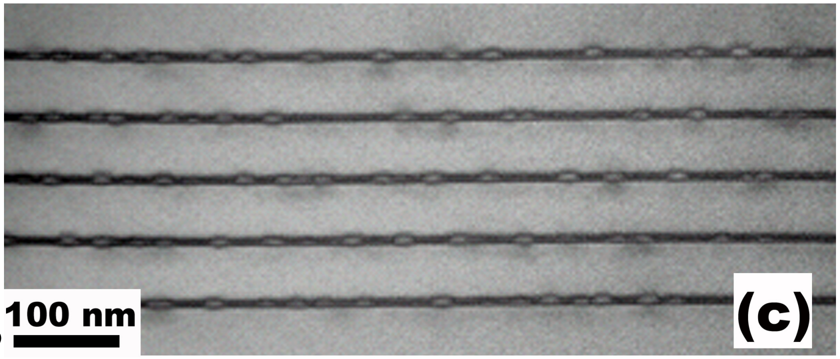
Origin of reduction of fine structure on annealing?

Flatter confinement potential, more 2D like (Bester et al PRB 67, 161306, 2003).

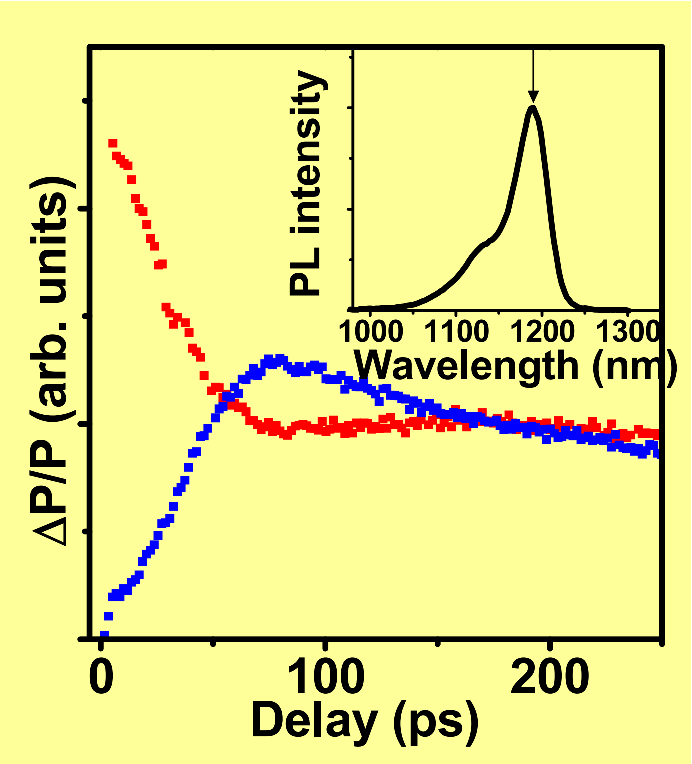
Possibly strain reduction on annealing, due to intermixing, reduces strain

DWELL (dot in a well, InAs dot in strain reducing InGaAs QW) show small splitting (~16μeV)

Reduced strain – reduced piezoelectric field – reduced fine structure splitting

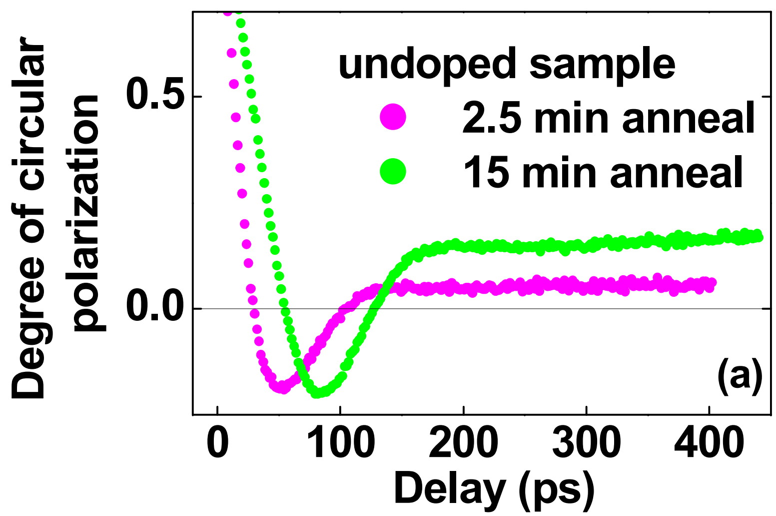


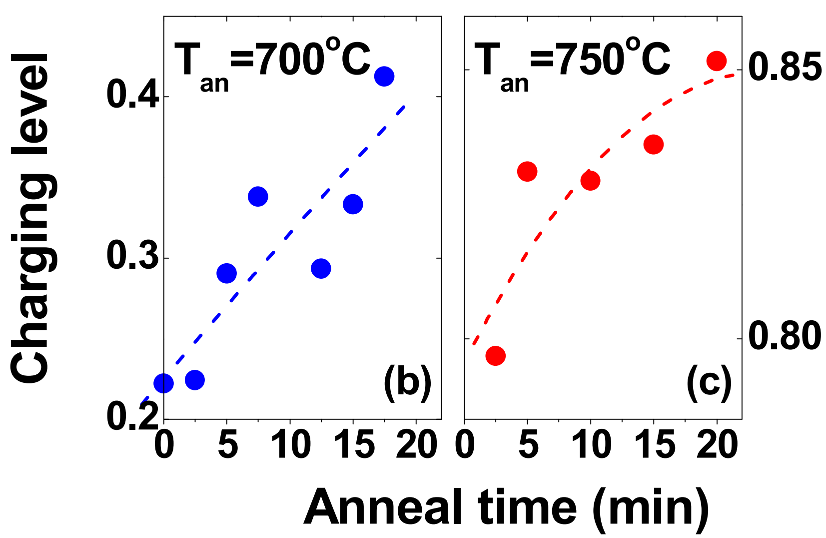
1.3 μm DWELLs



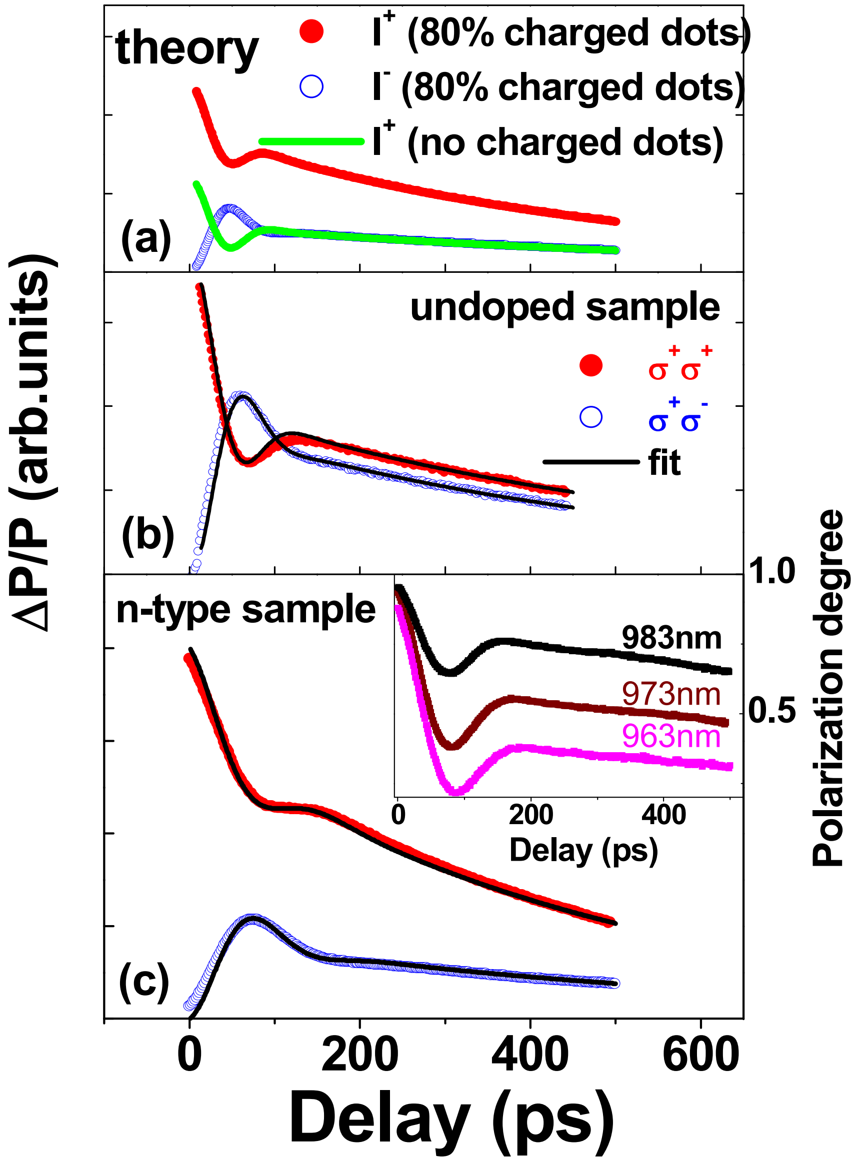
2. Second effect of annealing:

Fraction of charged dots increases (from 20 to 40% in 'undoped' samples)





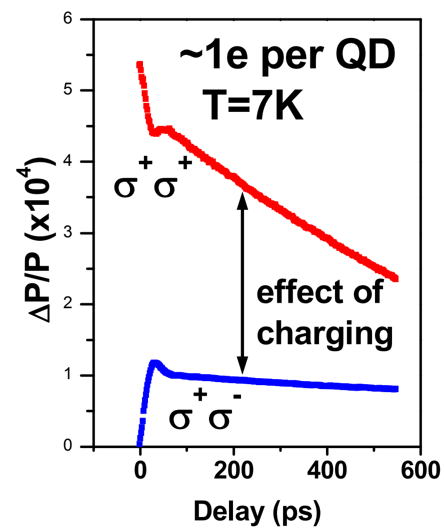
Deliberately doped samples (nominally one electron per dot



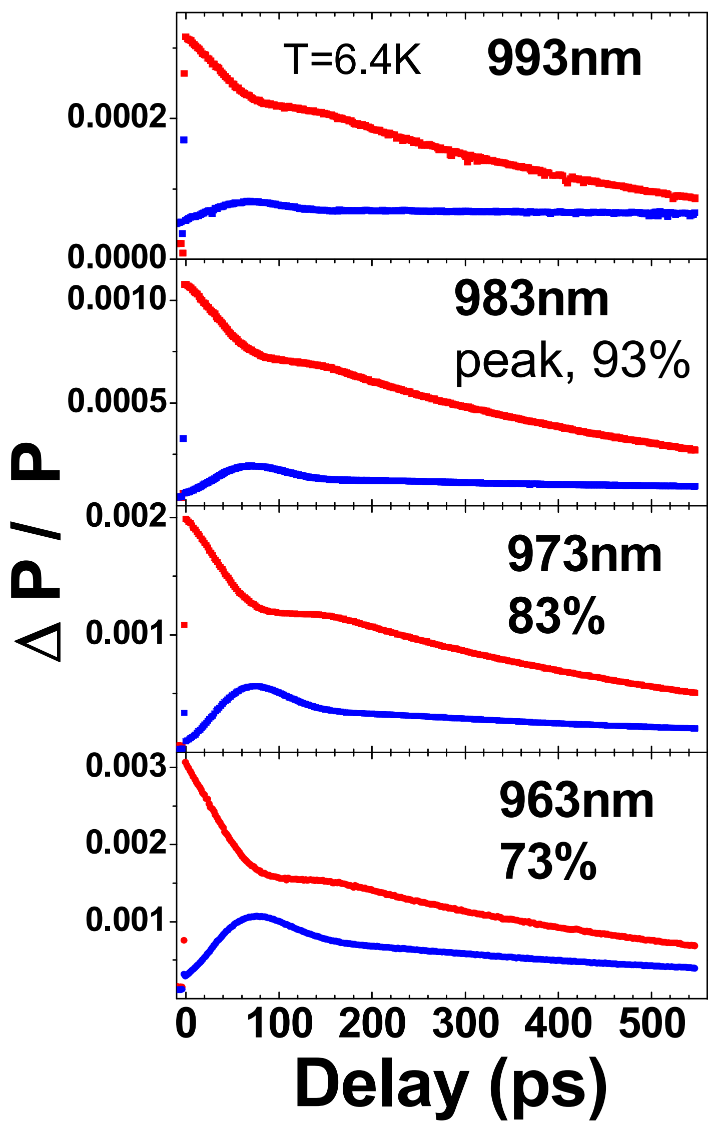
Large difference between σ^+ and σ^- probe signal levels in doped sample

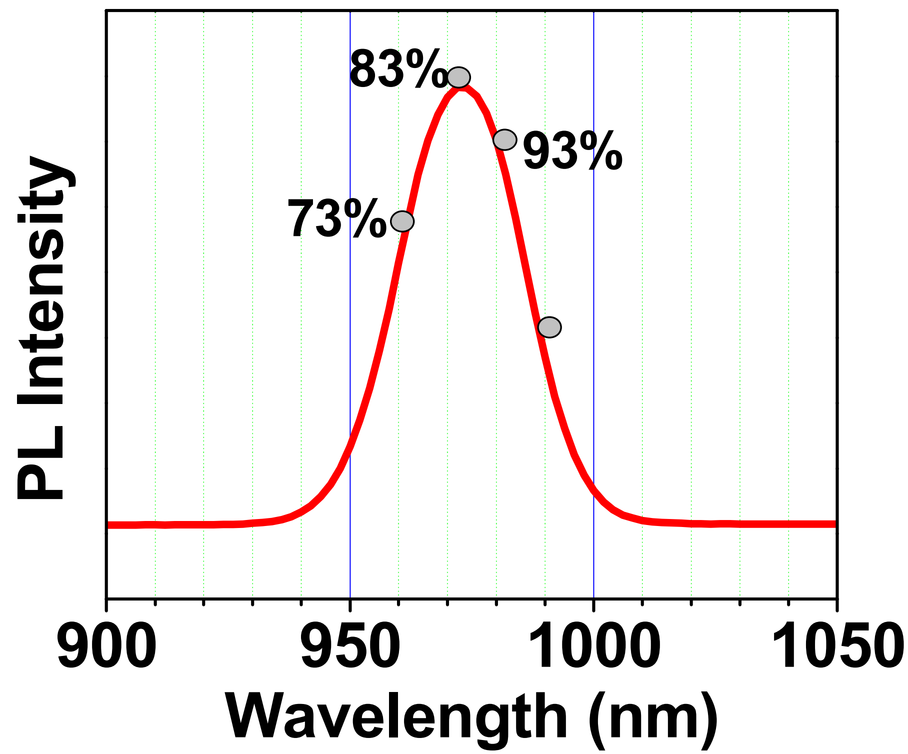
Large residual polarisation, >0.5

Corresponds to >80% of dots containing one electron



Variation of degree of polarisation and hence charged fraction with wavelength





Conclusions

1. Coherent and incoherent spin-related phenomena in pump-probe measurements
2. Spin coherence relatively robust up to ~100K
3. Long lived polarisation (spin) memory due to charged dots
4. Marked effects of annealing. Surprisingly can vary fine structure splitting
5. Probe of doping profile of dots, with energy resolution