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An introduction to the coherent optical control of a quantum dot spin

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PART 1: ENERGY LEVELS OF SEMICONDUCTOR QUANTUM DOT

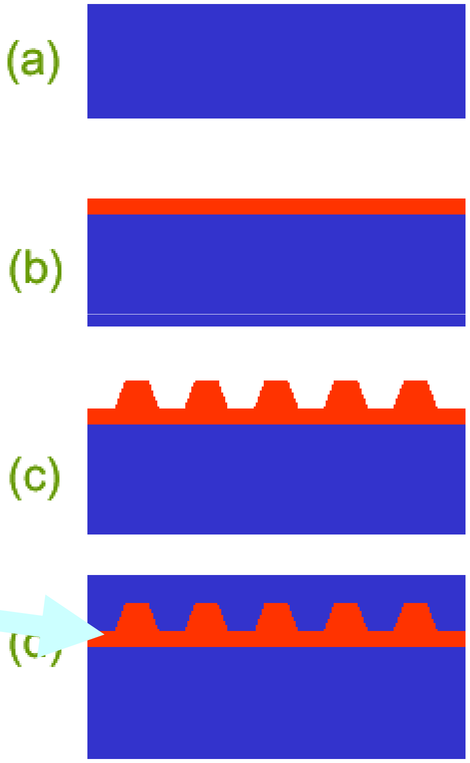
In(Ga)As self-assembled dots

Stranski-

Krastanow growth

🟥 InAs

🟦 GaAs



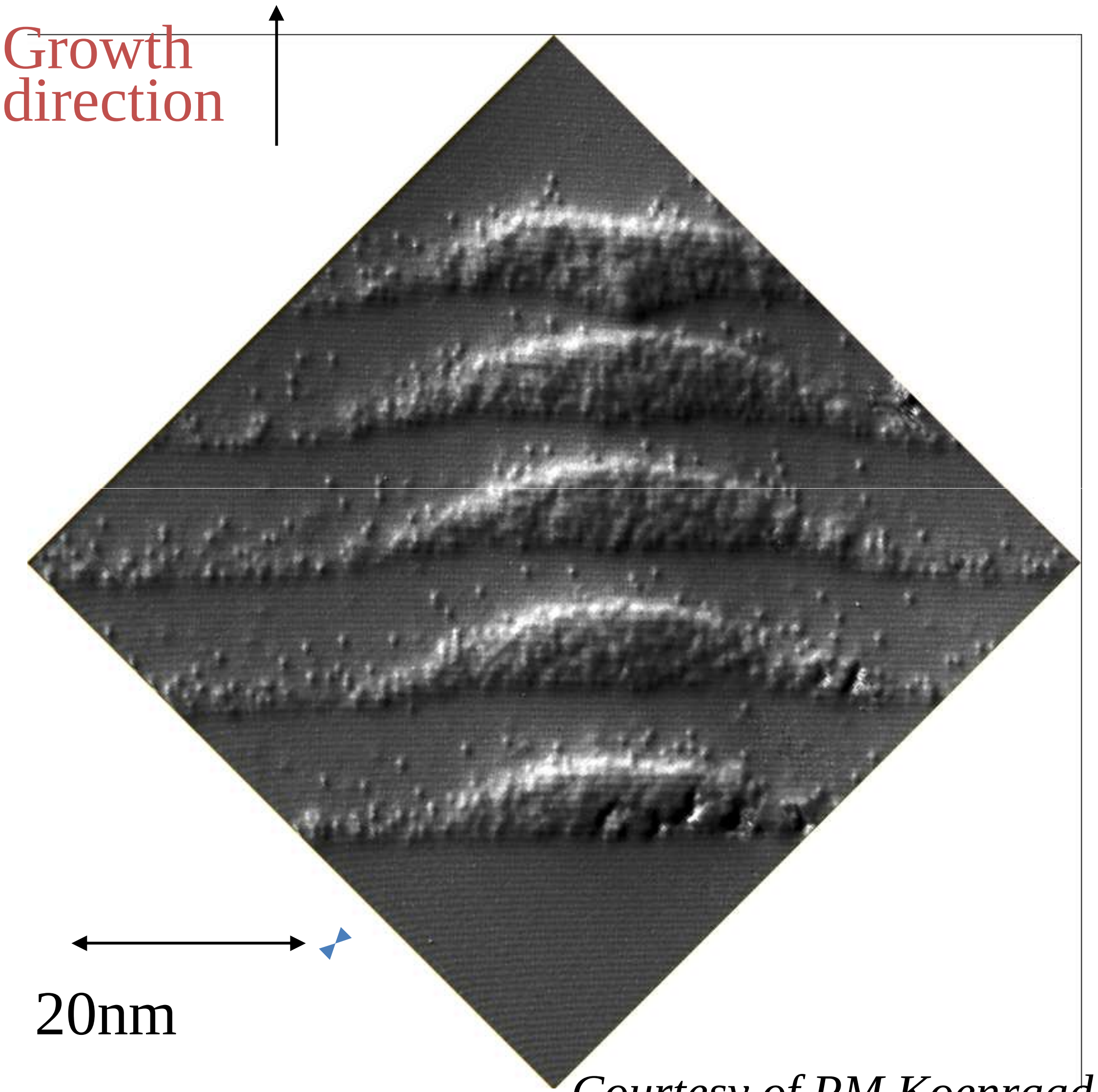
InAs-GaAs 7% lattice mismatch

Formation of quantum dots relieves strain

Note wetting layer

Embedded in crystal matrix – like any other semiconductor laser or light emitting diode

XSTM image of stack of InAs dots



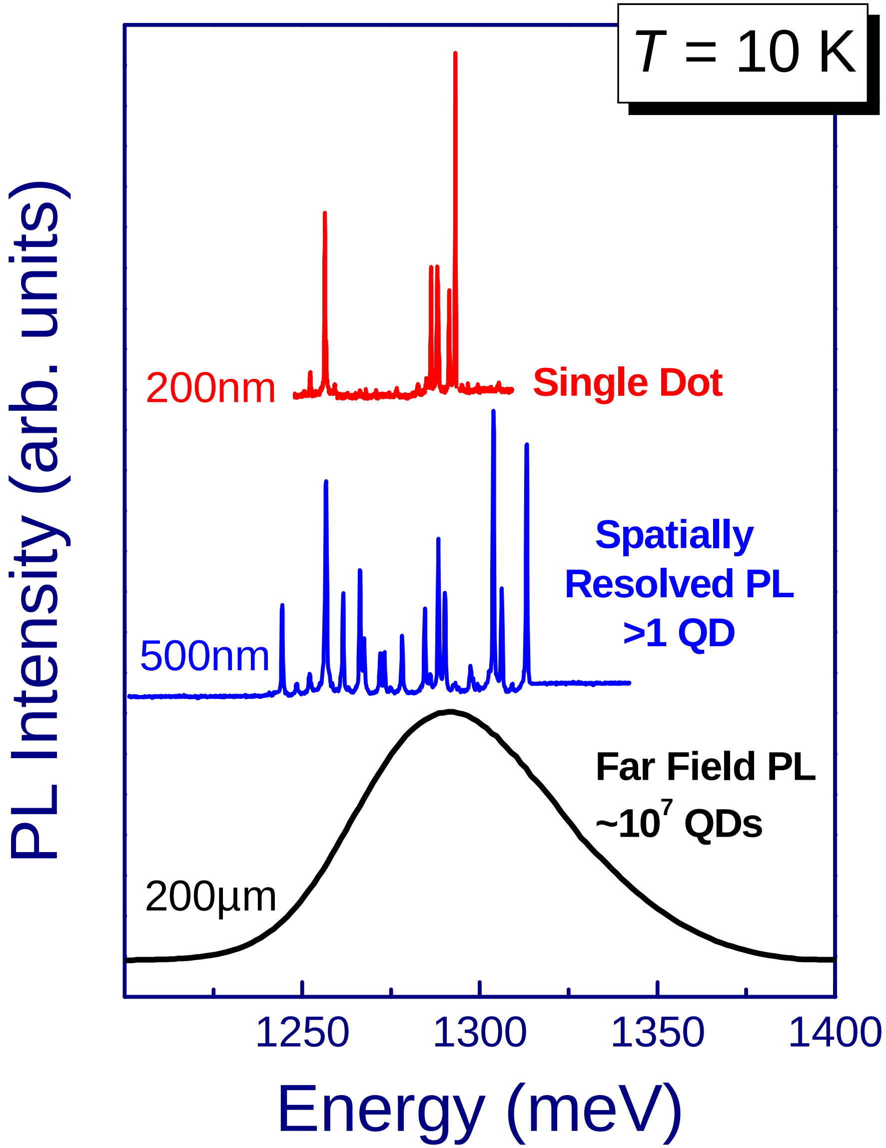
5 vertically aligned InAs quantum dots

Dots nucleate in strain field of preceding layer

Scanning tunnelling microscope image

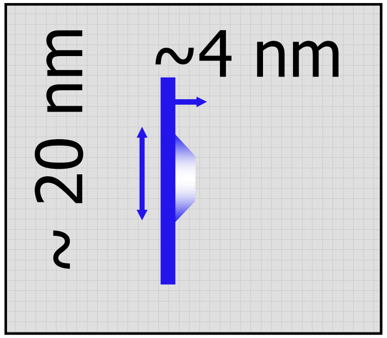
Courtesy of PM Koenraad, TU Eindhoven:

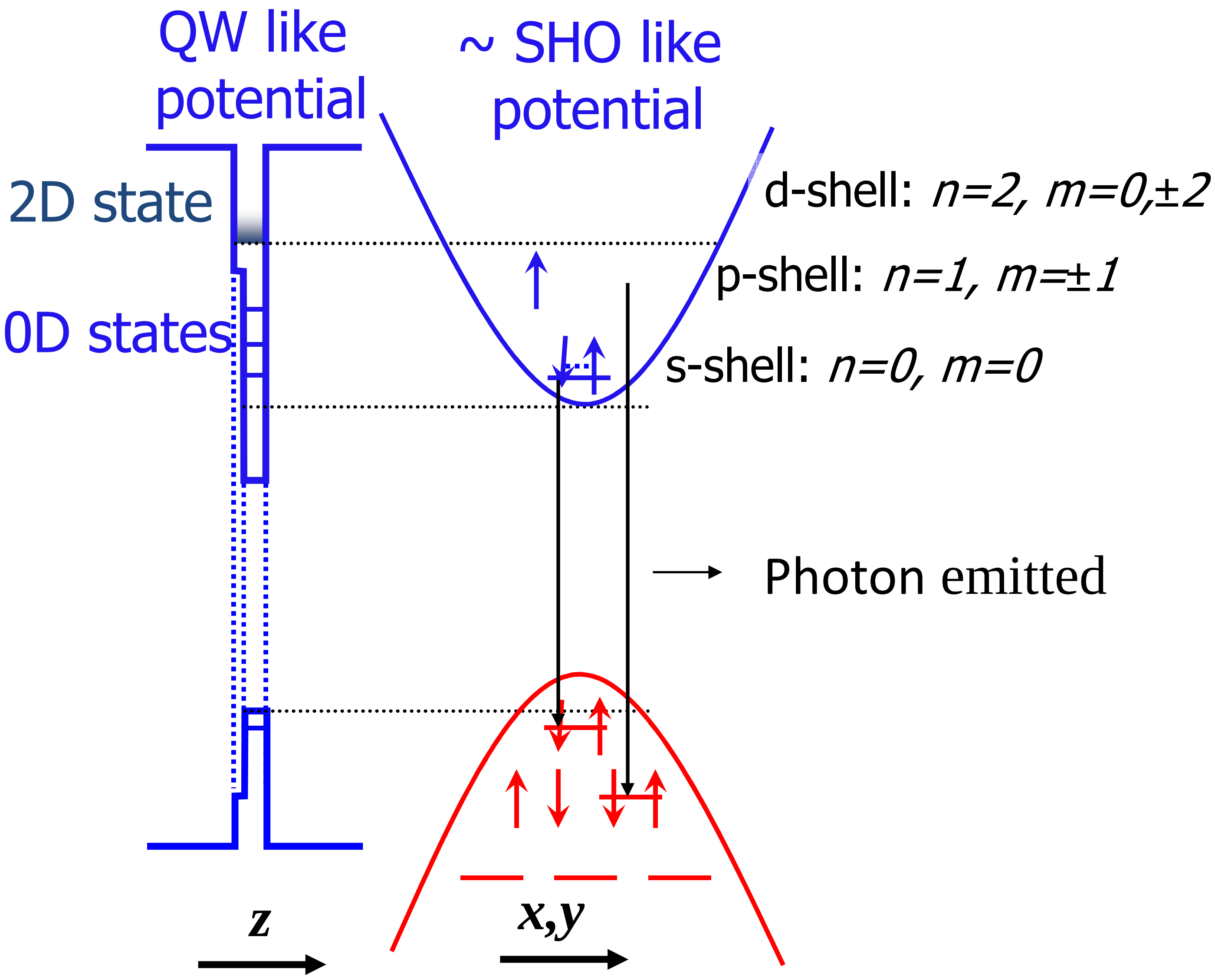
Spatially resolved PL



- Single dots may be optically isolated using apertures or mesas with ~400nm size.
- Emission spectrum breaks up into very sharp lines:
  FWHM ~ 50-150µeV
- Ground (s-shell) and excited state (p-shell) emission observed

Energy level structure





<u>Level spacing</u>

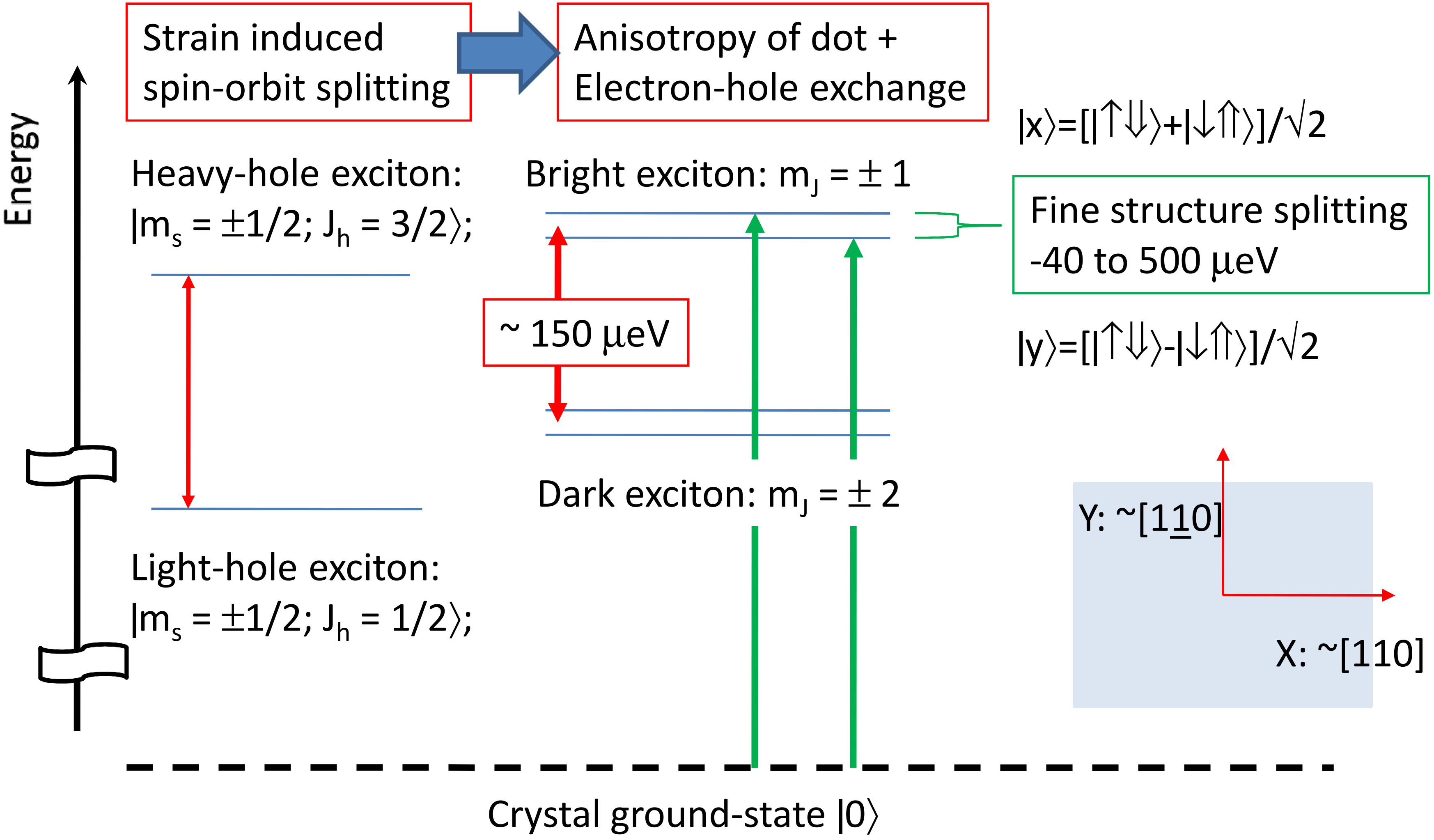
- Electrons:

  20-40meV

- Holes:

  10-30meV

Energy-level diagram of neutral exciton transitions



Strain induced spin-orbit splitting

Anisotropy of dot + Electron-hole exchange

Energy

Heavy-hole exciton:
$|m_s = \pm 1/2; J_h = 3/2\rangle$;

Light-hole exciton:
$|m_s = \pm 1/2; J_h = 1/2\rangle$;

Bright exciton: $m_J = \pm 1$

~ 150 μeV

Dark exciton: $m_J = \pm 2$

$|x\rangle = [|\uparrow\Downarrow\rangle + |\downarrow\Uparrow\rangle]/\sqrt{2}$

Fine structure splitting
-40 to 500 μeV

$|y\rangle = [|\uparrow\Downarrow\rangle - |\downarrow\Uparrow\rangle]/\sqrt{2}$

Y: ~[1$\underline{1}$0]

X: ~[110]

Crystal ground-state $|0\rangle$

Summary part 1

- Quantum dots are 3D finite quantum wells
- Discrete energy-levels
- Have an atom-like light-matter interaction

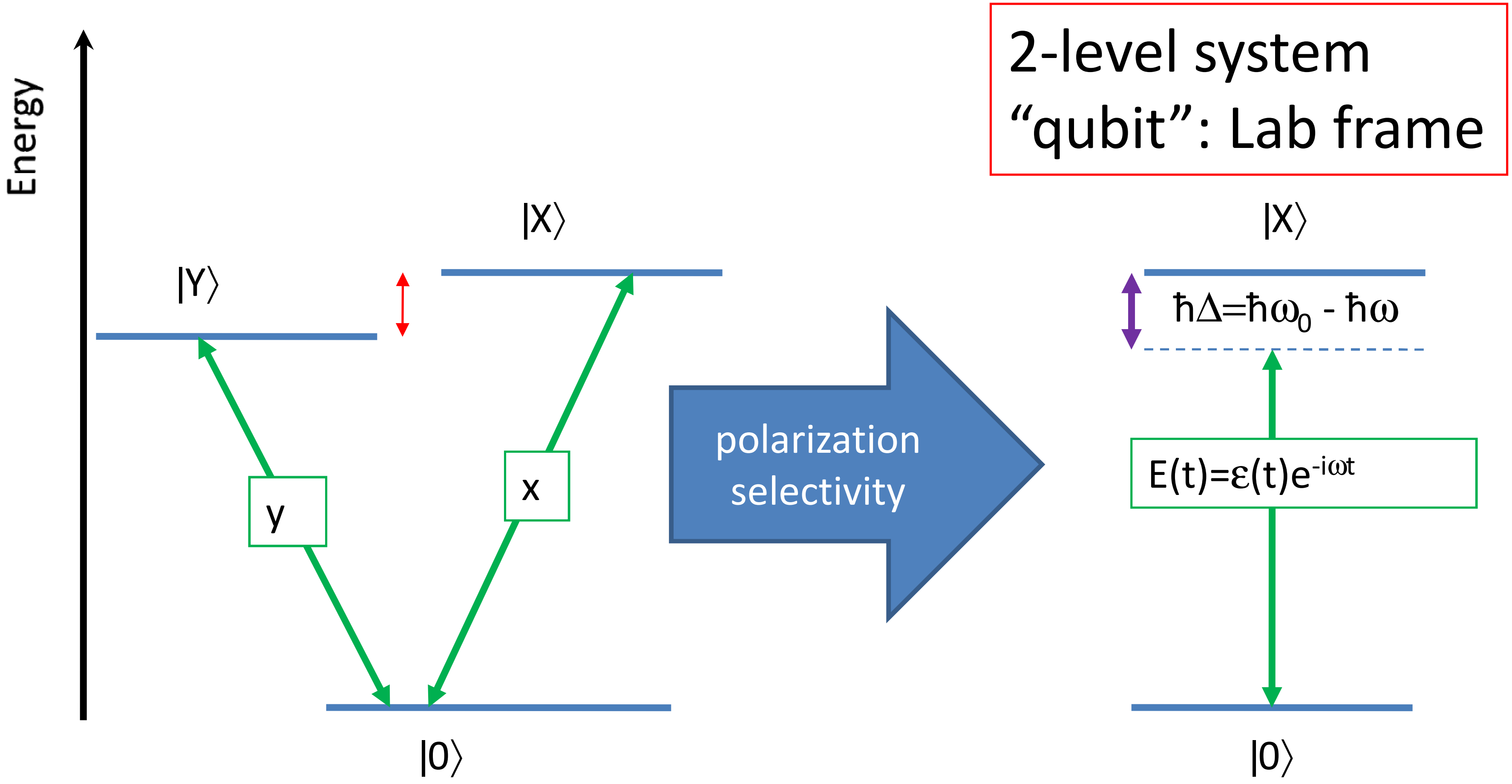
PART 2: COHERENT DYNAMICS OF AN IDEAL 2-LEVEL SYSTEM DRIVEN BY A LASER

What is coherent control?

- H= H(system) + H(control)

Simplified energy-level diagram of neutral exciton transition

2-level system
“qubit”: Lab frame



Energy

$|Y\rangle$

$|X\rangle$

y

x

$|0\rangle$

polarization selectivity

$|X\rangle$

$\hbar\Delta = \hbar\omega_0 - \hbar\omega$

$E(t) = \varepsilon(t)e^{-i\omega t}$

$|0\rangle$

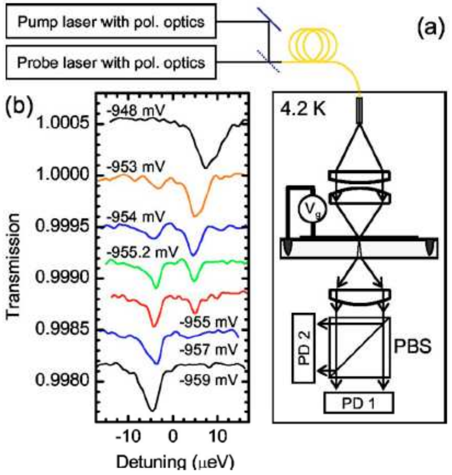
laser field oscillates pretty fast ~ 330 THz

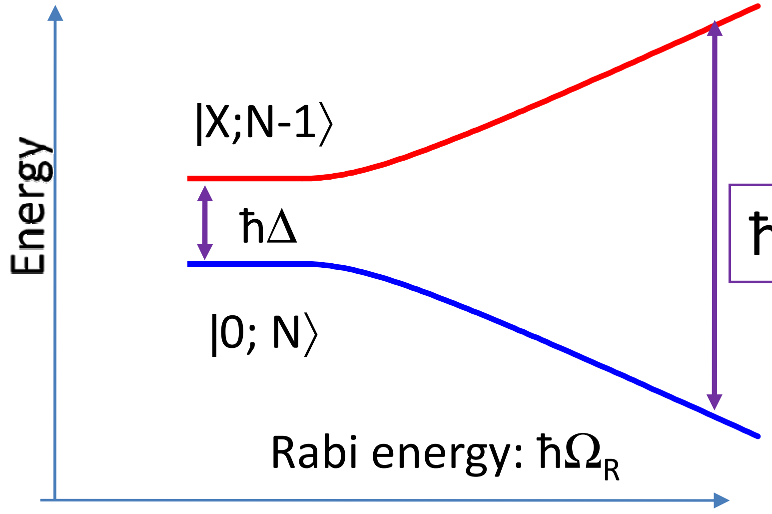
Energy eigenstates of optically coupled 2-level system in the dressed states, or rotating frame picture

|exciton; photon number⟩

$$H = \frac{\hbar}{2}\begin{pmatrix} -\Delta(t) & \Omega_R(t) \\ \Omega_R^{*}(t) & \Delta(t) \end{pmatrix} \begin{matrix} |0;\ N\rangle \\ |X;\ N-1\rangle \end{matrix}$$

$\hbar\Omega_R(t) = \mu\varepsilon(t)/2$; μ = optical dipole mome





Rabi energy: $\hbar\Omega_R$

$|\alpha\rangle \rightarrow [|0;N\rangle + |X;N-1\rangle]/\sqrt{2}$

$\hbar\Omega_{eff} = \hbar\sqrt{(\Delta^2 + |\Omega|^2)}$

$|\beta\rangle \rightarrow [|0;N\rangle - |X;N-1\rangle]/\sqrt{2}$

M. Kroner,
APL 92 031108
(2008).

Rabi oscillation: beat between two dressed states, under resonant excitation Δ =0

At t=0, initially dot is in exciton ground-state:

$|0;N\rangle=[|\alpha\rangle+|\beta\rangle]/\sqrt{2} \rightarrow$

This is a superposition of the dressed states, which beats:

$$[\exp(i\Theta(t)/2)|\alpha\rangle+\exp(-i\Theta(t)/2)|\beta\rangle]/\sqrt{2} \; =$$

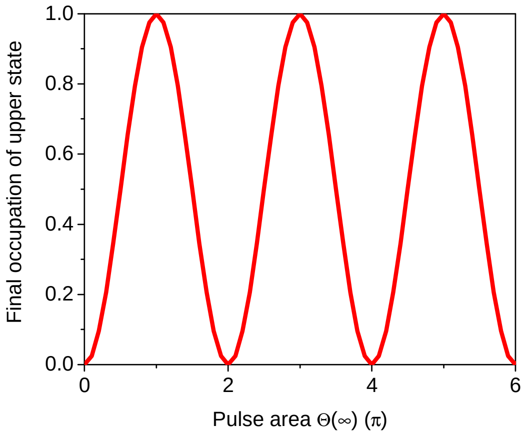
Causing an oscillation in the occupation of the upper state:

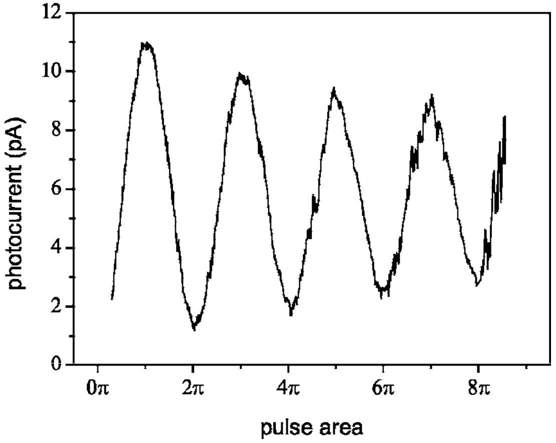
$$\cos[\Theta(t)/2]|0;N\rangle-i.\sin[\Theta(t)/2]|X;N-1\rangle$$

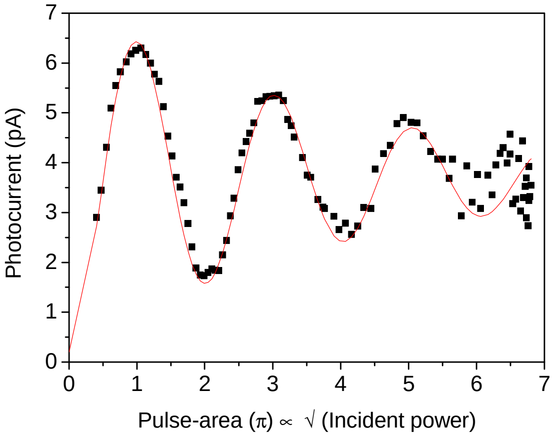
Where

$\Theta(t) = \int^{t}_{0} \Omega_R(s)ds$, and $\Theta(\infty)$ is the pulse-area

Rabi oscillations: final occupation of upper state “|X〉” versus pulse-area







Experimental data:

Calculation for ideal 2-level atom

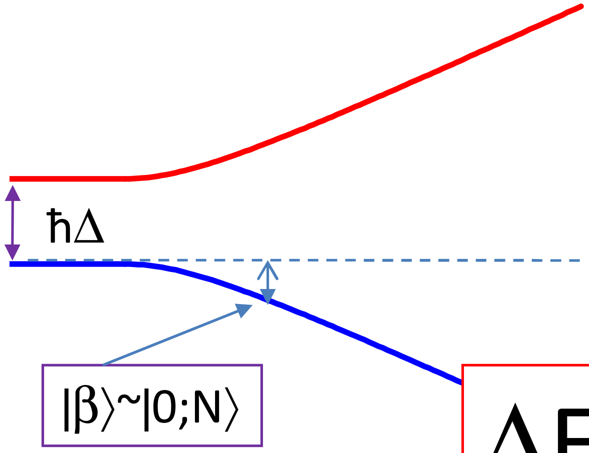
T = 4.2 K
S. Stufler, et al PRB 121301R (2005).

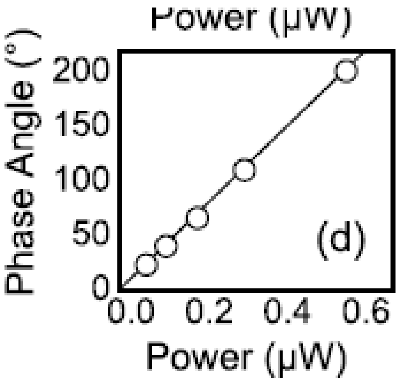
T = 15 K
Sheffield

In general Rabi oscillations are damped for reasons that will be discussed later

Single qubit operation to control occupations of upper and lower energy states

AC Stark-shift: excitation far from resonance $\Delta >> \Omega_R$, i.e. far from resonance Rabi oscillation



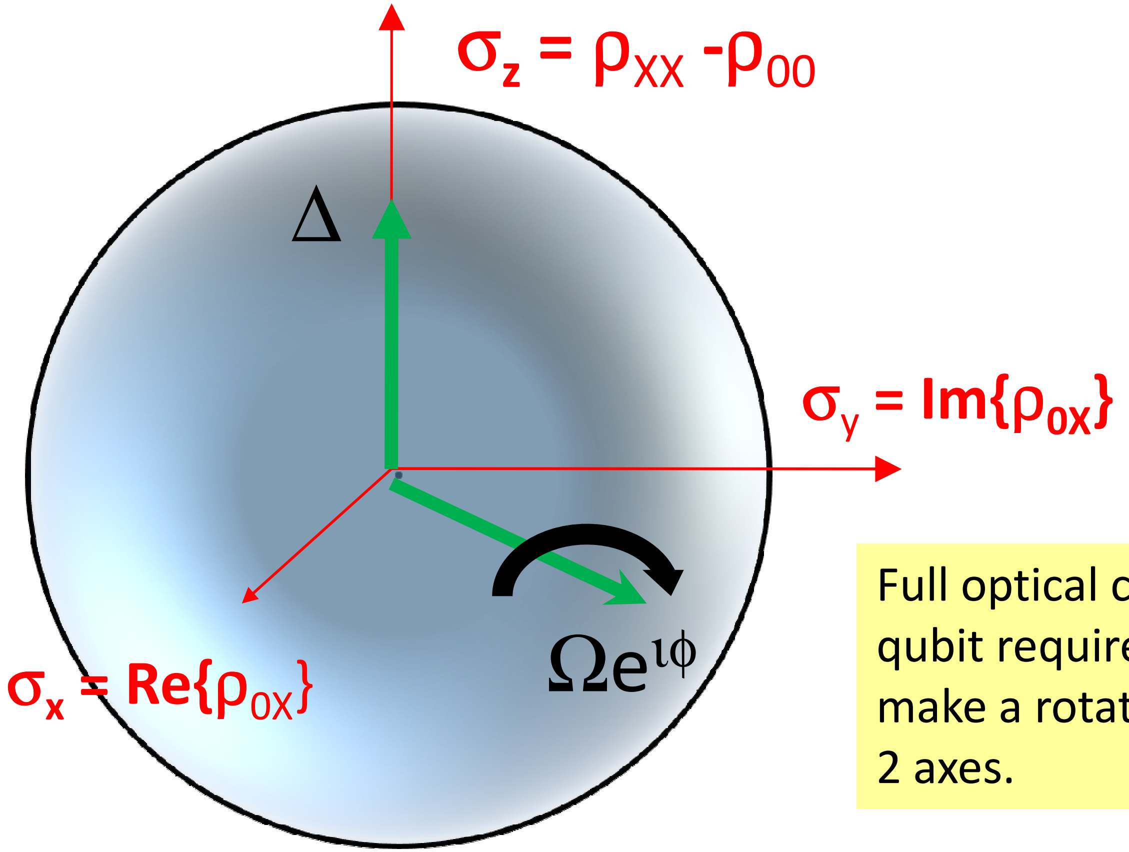


Unold, PRL 92 157401 (2002).
AC-Stark shift ∝ power

$$\Delta E_{AC} = \hbar[\Delta - \sqrt{(\Delta^2 + \Omega_R^2)}]/2$$
$$\sim -\hbar\Omega_R^2 /4\Delta$$

Application of a laser pulse far from resonance results in weak mixing of the 0-X states,
but does result in a shift in the energy
This can be used to control the relative phase of the qubit.

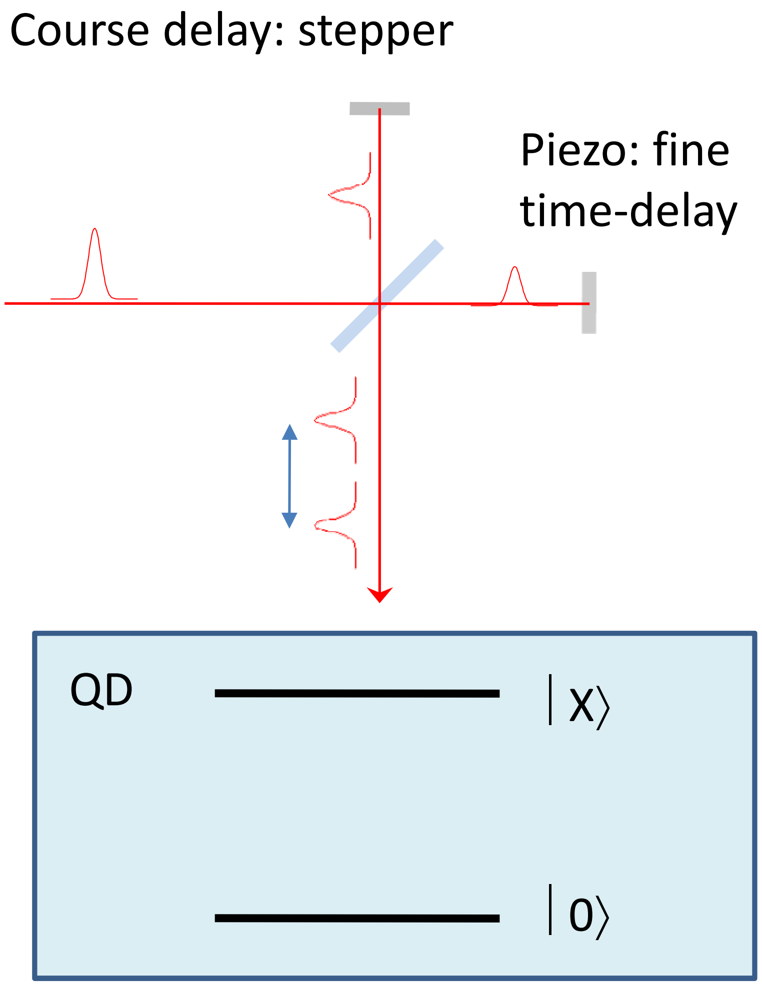
Bloch-sphere representation of the density matrix $\rho_{ij} = |i\rangle\langle j|$ of a 2-level system (qubit)



Full optical control of qubit requires ability to make a rotation about 2 axes.

$$H = \Delta\sigma_z + \mathrm{Re}\{\Omega\}\sigma_x + \mathrm{Im}\{\Omega\}\sigma_y$$

Two-pulse quantum (Ramsey) interference (1)

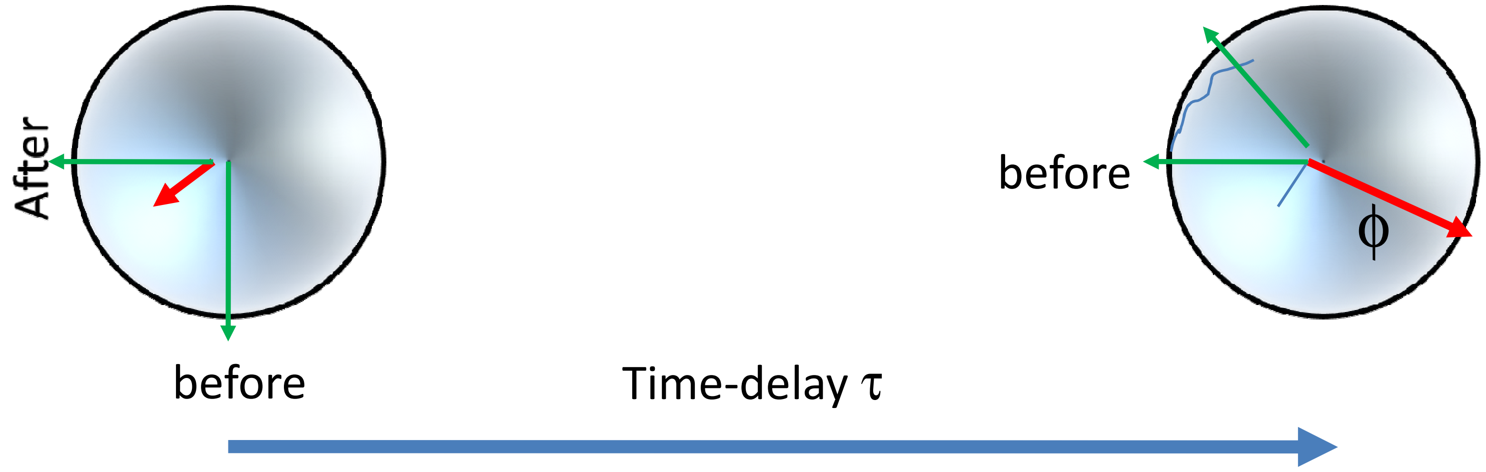


Interferometer creates two pulses with interferometrically stable time-delay

$$\Omega(t) = f(t)e^{\iota\omega t} + f(t-\tau)e^{\iota\omega(t-\tau)}$$

phase of pulse defines the rotation axis

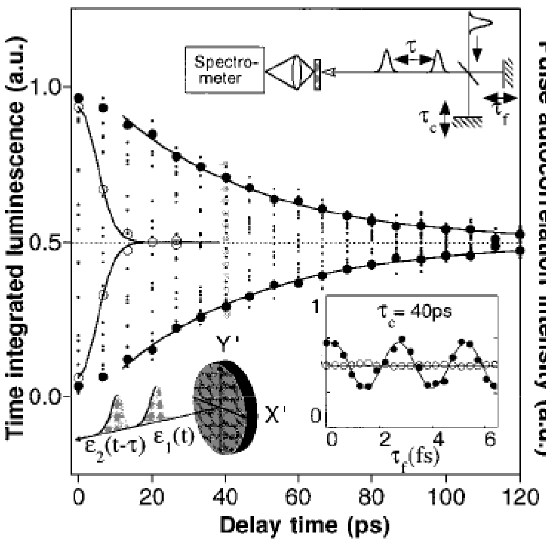
Two pulse quantum (Ramsey) interference, (2)



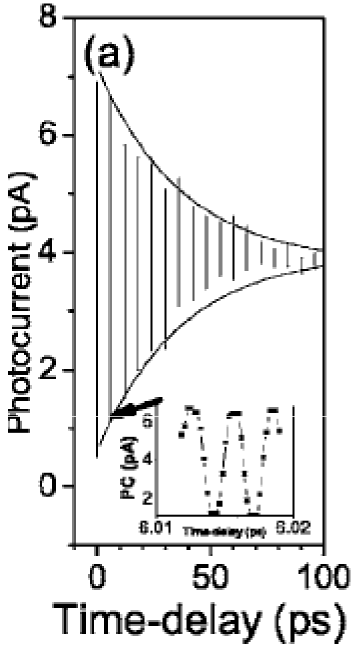
At time t=0, first pulse
Rotates Bloch-vector by angle π/2
About x-axis

At time t= τ, second pulse
Rotates Bloch vector by angle π/2
About axis: **x**. cos(ϕ) + **y**.sin(ϕ)
Where ϕ = ωτ

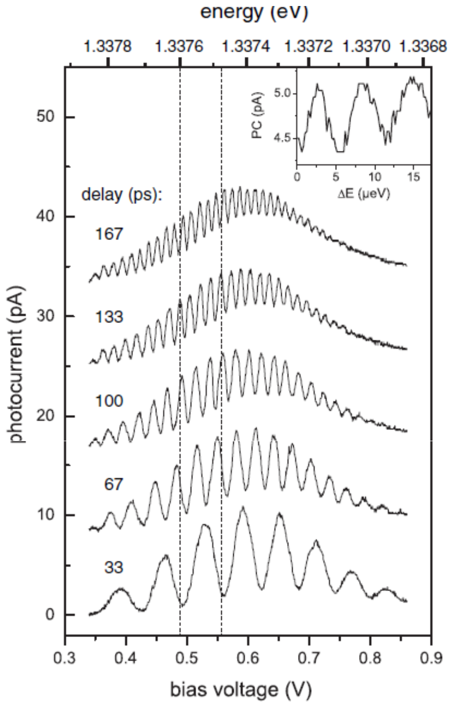
Examples of quantum interference data



N. H. Bonadeo, Sci 282 1473 (1998).



R. S. Kolodka, PRB 75 193306 (2007).



S. Stufler, PRL 96 037402 (2006).

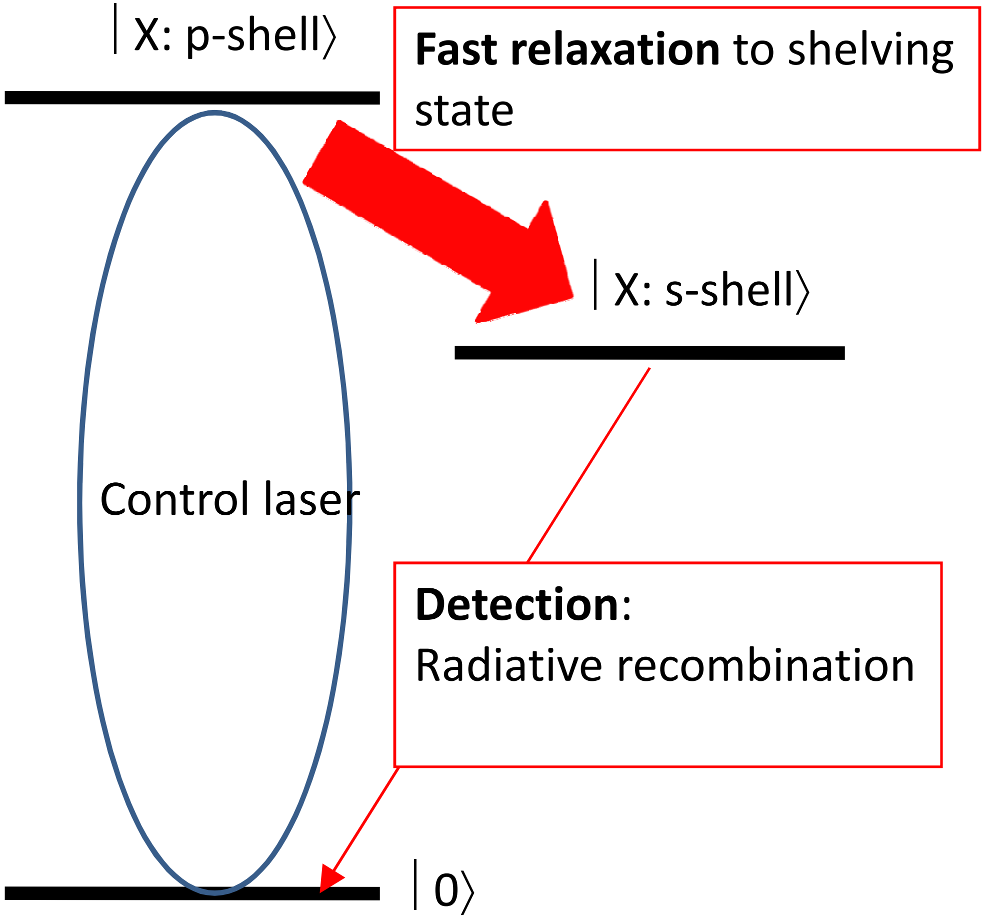
Part 2 summary

- A qubit can be represented as a point on a Bloch sphere
- Full control requires ability to rotate about two axes
- A laser pulse couples the upper and lower states, causing a rotation about axis in xy-plane with angle determined by phase of electric-field
- Single pulse: Rabi oscillation
- Double pulse: Ramsey interference
- The detuning of the laser pulse (energy-splitting) can be used to “tip” the rotation axis to give a z-component to the rotation: AC-Stark effect

PART 3: EXPERIMENTAL APPROACHES

So how do you separate a weak (< one photon) signal from the reflected laser pulses of the same wavelength, and polarization as the signal?

P-shell control



Fast relaxation to shelving state

Detection:
Radiative recombination

PRO: spectrally filter the control laser from the signal
CON: p-shell states are short lived ~ 30 ps

Examples:
Bonadeo, Sci 282 1473 (1998).
Htoon, PRL 88 087401 (2002).
Besombes, PRL 90 257402 (2003).
Kamada, PRL 87 246401 (2001).

Four-wave mixing

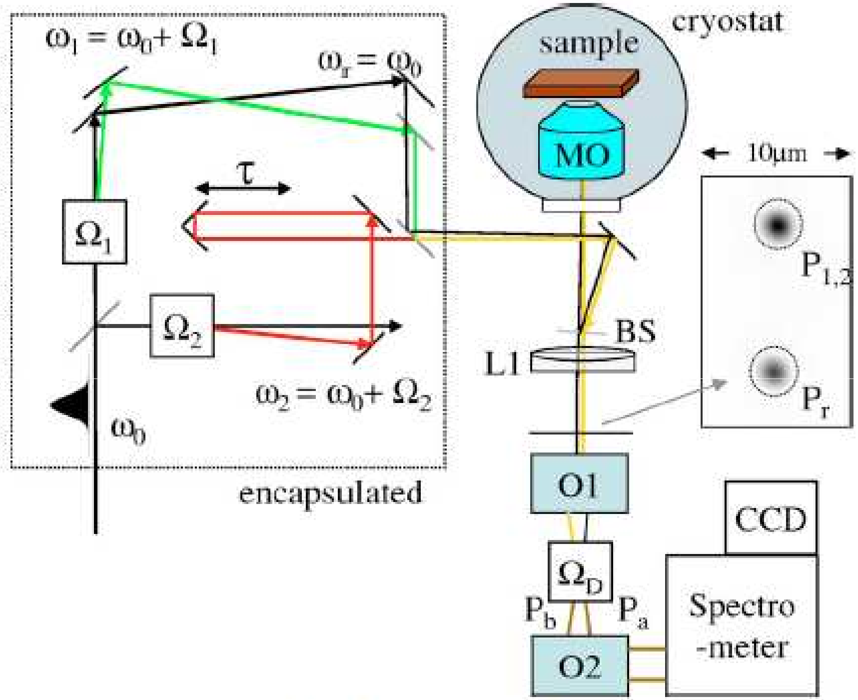


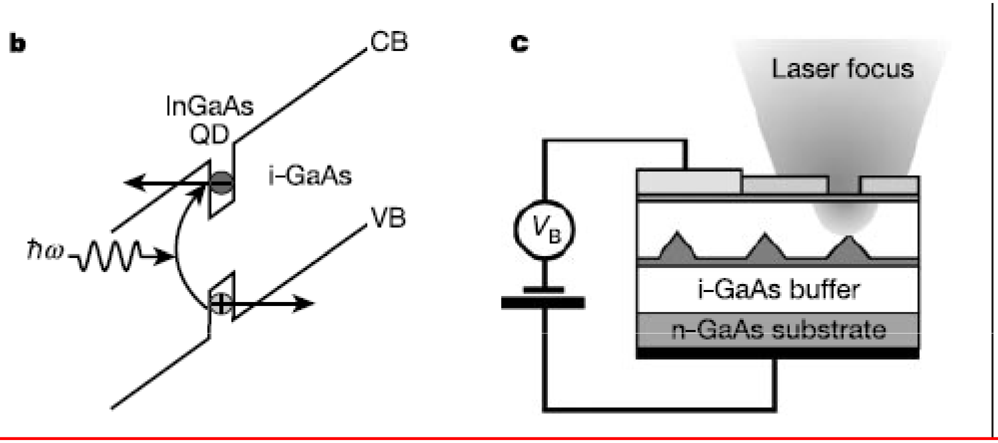
Fig. 1. (Color online) Schematic of the experimental setup.

PRO: Powerful technique can retrieve full information on dot dynamics; does not require custom devices
CON: Complicated, weak signals, only recently have measurements on InAs dots been achieved

Examples: X. Li, Sci 301 809 (2003); Langbein and Patton, PRL 95 017403 (2005).

From: Langbein and Patton, Opt. Lett. 31 1531 (2006).

Photocurrent detection

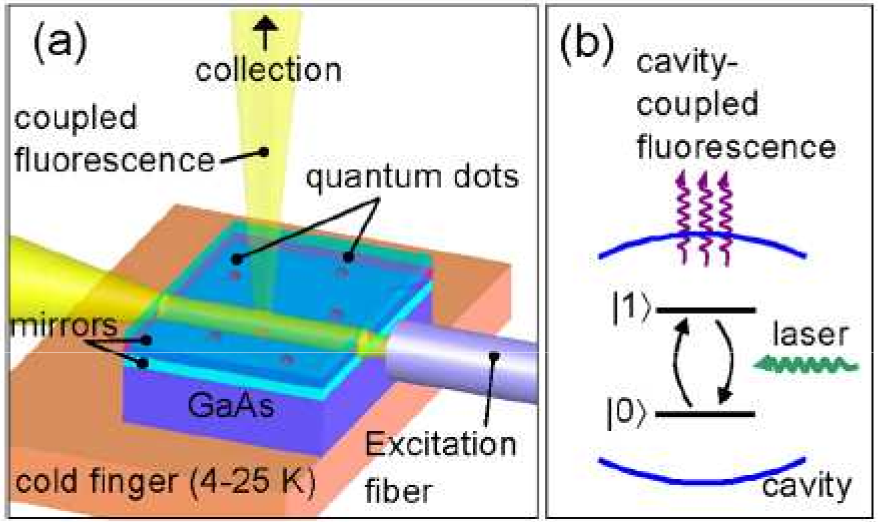


From: Zrenner, Nature 418 612 (2002).

Absorption of photon creates electron-hole pair in dot, which under applied electric-field tunnels from the dot the resulting in a photocurrent

PRO: Highly efficient quantitative detection, fast measurement times
CON: Destructive measurement, coherence times limited by tunneling
Further examples: Stufler, PRL 96 037402 (2006); Ramsay PRL 100 197401 (2008); Takagi, Optics Exp. 16 13949 (2008).

Resonant fluorescence



A. Muller, PRL 99 187402 (2007).

Dot in a cavity is excited transverse to the cavity direction ⇒ extremely good separation of fluorescence and excitation laser, even if at same frequency

PRO: Can access new physics (e.g. Mollow triplet, Raman scatterring); Single photon output, creates new possibilities for measurement based QIP, and integration with linear optics QIP schemes etc
CON: Polarization is a problem,

Other examples: E. B. Flagg, Nat Phys. , E. B. Flagg, PRL 10; K. Kuroda, APL, 90 051909 (2007); R. Melet, PRB 78 073301 (2008);

PART 4: COHERENT OPTICAL CONTROL OF SPINS

Motivation, finding a good solid-state qubit

Exciton

PRO: strong optical dipole, allowing optical control on ps timescale

CON: short coherence times < 1 ns

Spin

PRO: extremely long coherence times: T_1 ~ 20 ms, $T_2 > 10\ \mu s$ for electron

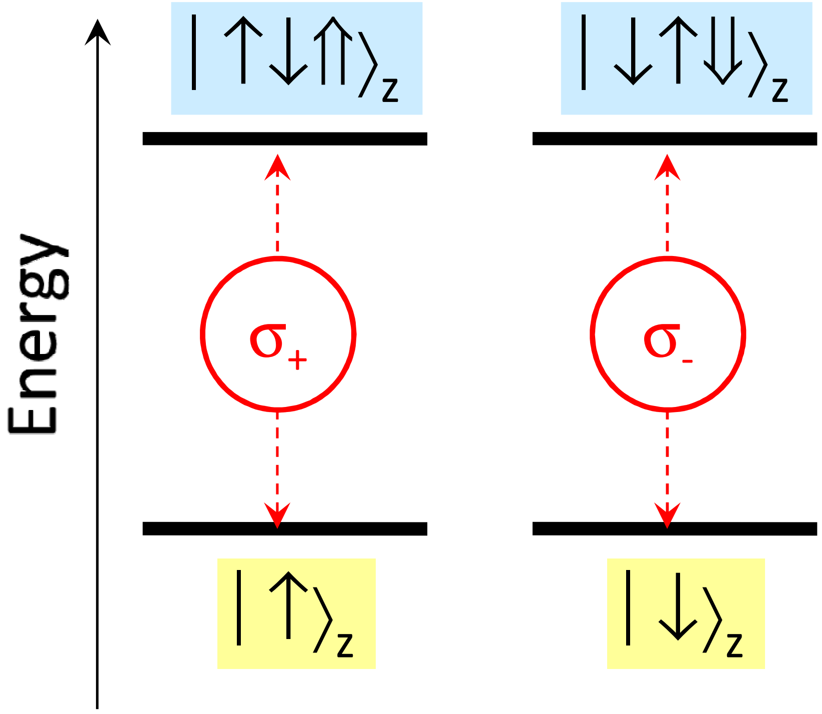
CON: small Zeeman splitting, giving slow control

“Best of both solution”: Use charged excitons for ps control of long-lived spin qubit

The challenge

- Initialization: unlike an exciton qubit, the spin is not in a well-defined initial state
- Detection: need to time-resolve a single spin
- Control: a bit more complicated than exciton control

Energy-level diagram of electron-trion system: no B-field



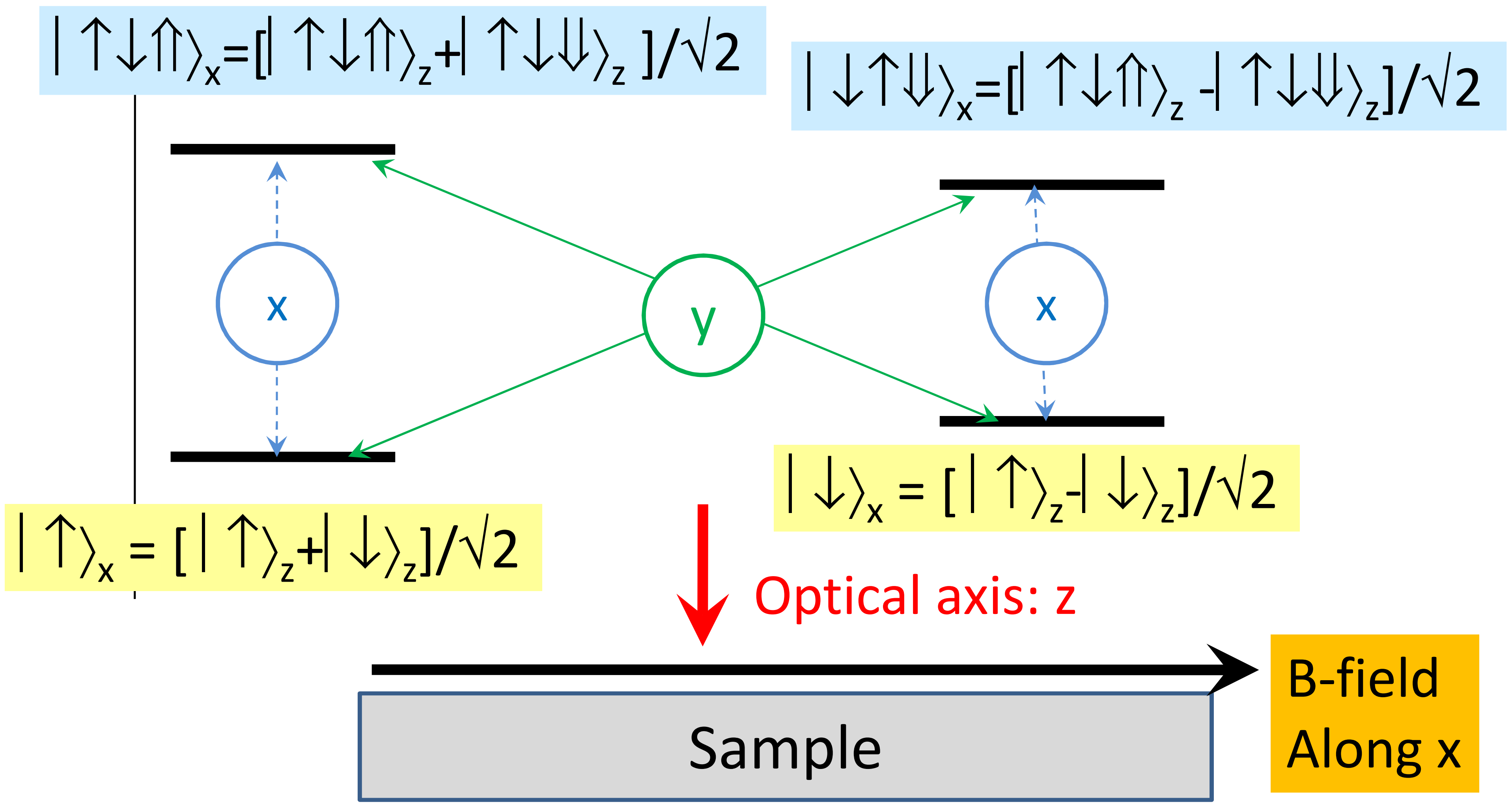
Circular-polarization can impart a phase-shift on target spin

⇒ z-rotation on Bloch sphere

⇒ However, need coupling between spins to do a x or y rotation.

No B-field: 2 independent 2-level atoms

Simplest solution: Voigt geometry B-field (along sample)



$|\uparrow\downarrow\Uparrow\rangle_x=[|\uparrow\downarrow\Uparrow\rangle_z+|\uparrow\downarrow\Downarrow\rangle_z]/\sqrt{2}$

$|\downarrow\uparrow\Downarrow\rangle_x=[|\uparrow\downarrow\Uparrow\rangle_z-|\uparrow\downarrow\Downarrow\rangle_z]/\sqrt{2}$

$|\downarrow\rangle_x=[|\uparrow\rangle_z-|\downarrow\rangle_z]/\sqrt{2}$

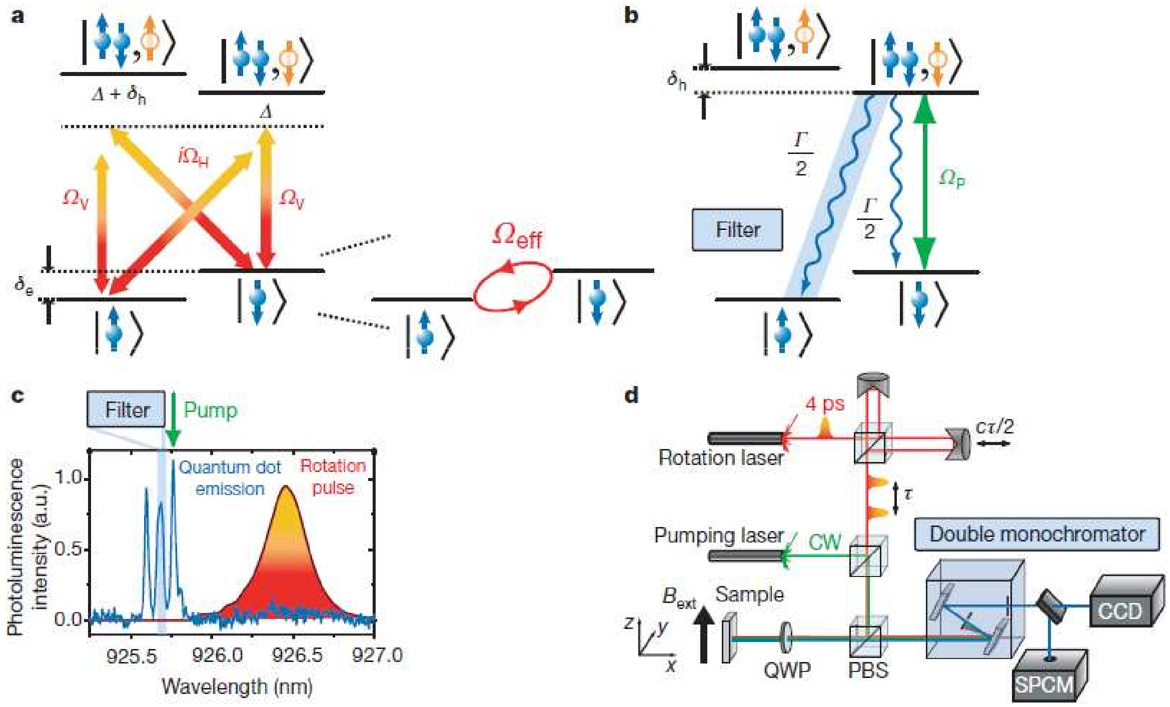
$|\uparrow\rangle_x=[|\uparrow\rangle_z+|\downarrow\rangle_z]/\sqrt{2}$

Optical axis: z

Sample

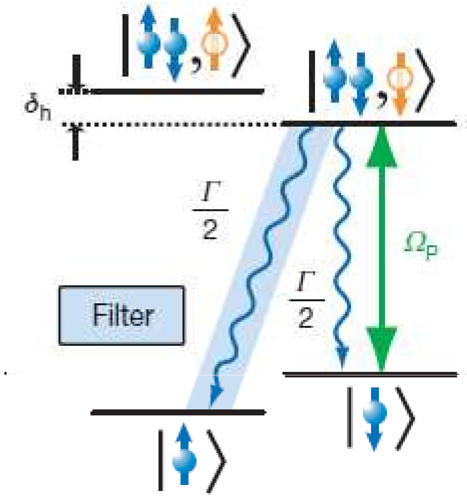
B-field
Along x

Example 1: Coherent optical control of a single electron spin: Press et al, Nature 456 218 (2008), using an InAs dot



Note: spin is aligned along B-field

Press Nature 418 232 (2008): initialization and detection

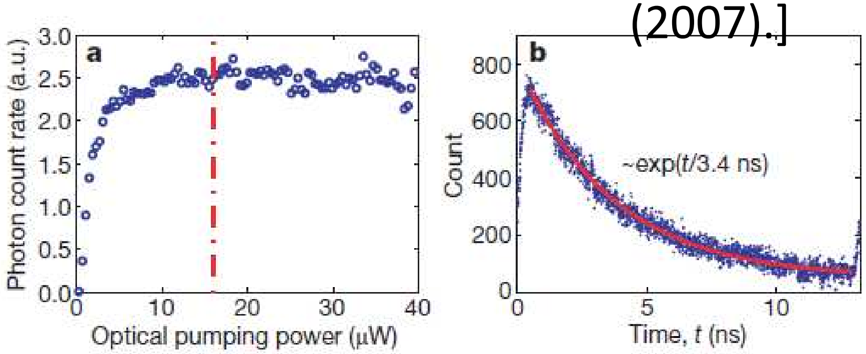


CW laser with circular polarization drives $|\downarrow\rangle \leftrightarrow |\uparrow\downarrow\Downarrow\rangle$ transition.

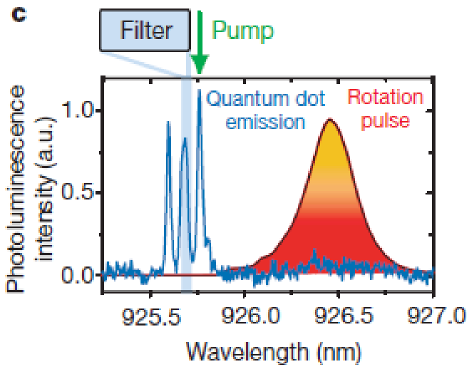
Λ-transition: Stimulated Raman scattering results in inelastic scattering of the photon, with a spin-flip

Detection of Raman scattered photon $\Rightarrow$ dot was in spin $\downarrow$ state,

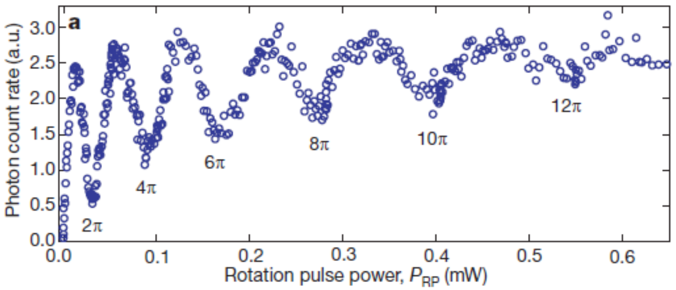
Initialization: if the spin is not flipped laser continues to drive transition, if spin is flipped dot is no longer in resonance with laser [see also: X. Xu, PRL 99 097401 (2007).]



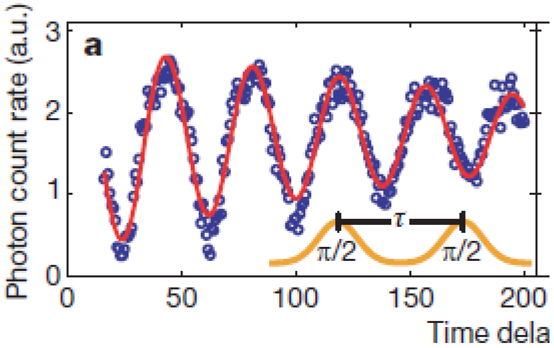
Press, Nature 418 232 (2008): coherent control of electron spin using AC-Stark shift



Circularly polarized laser pulse, far from resonance



“Rabi” oscillations
Z-rotation



“Ramsey interference”:
X-rotation

Initial state: $|\uparrow\rangle_x = [\,|\uparrow\rangle_z + |\downarrow\rangle_z\,] / \sqrt{2}$

Off resonant laser pulse puts phase shift on $|\downarrow\rangle_z$ due to AC-Stark effect

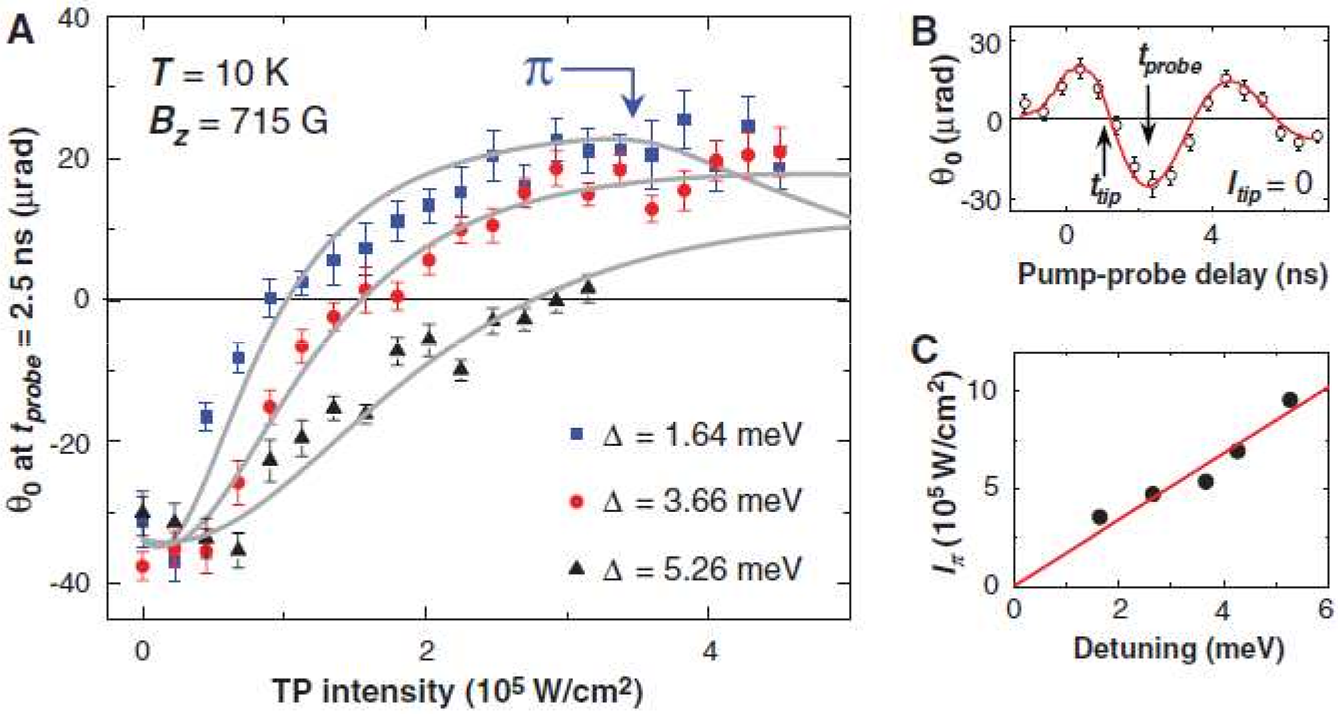
$$\Rightarrow |\psi\rangle \rightarrow [\,|\uparrow\rangle_z + e^{i\varphi}|\downarrow\rangle_z\,],\ \varphi \sim \int dt.\ \Omega^2/4\Delta$$

$$= \cos(\varphi/2)|\uparrow\rangle_x + i.\sin(\varphi/2)|\downarrow\rangle_x$$

Single pulse: rotates between spin up and down (x-basis), Rabi oscillation

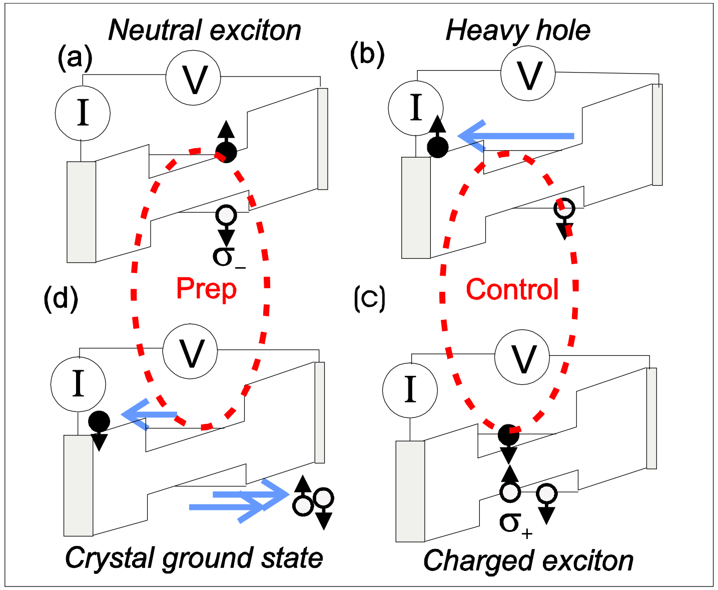
Two pulses: controls rotation axis, Ramsey interference

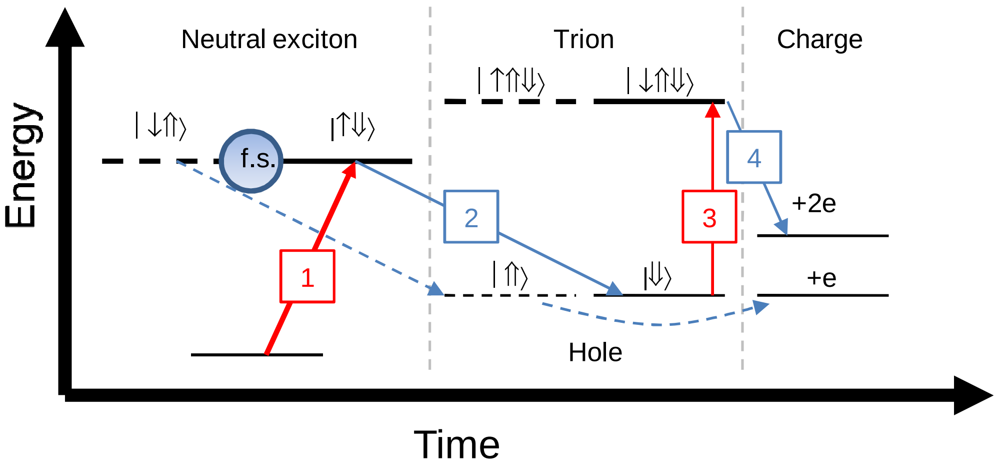
Example 2: Berezovsky et al, Sci 320 349 (2008), z-rotation of electron spin in GaAs interface dot, using AC-Stark effect, and a Kerr-rotation detection method



Rotation is strongly damped, are GaAs dots suitable for coherent optical control?

Example 3: Photocurrent approach, initialization by exciton ionization, Ramsay PRL 100 097401 (2008). No B-field

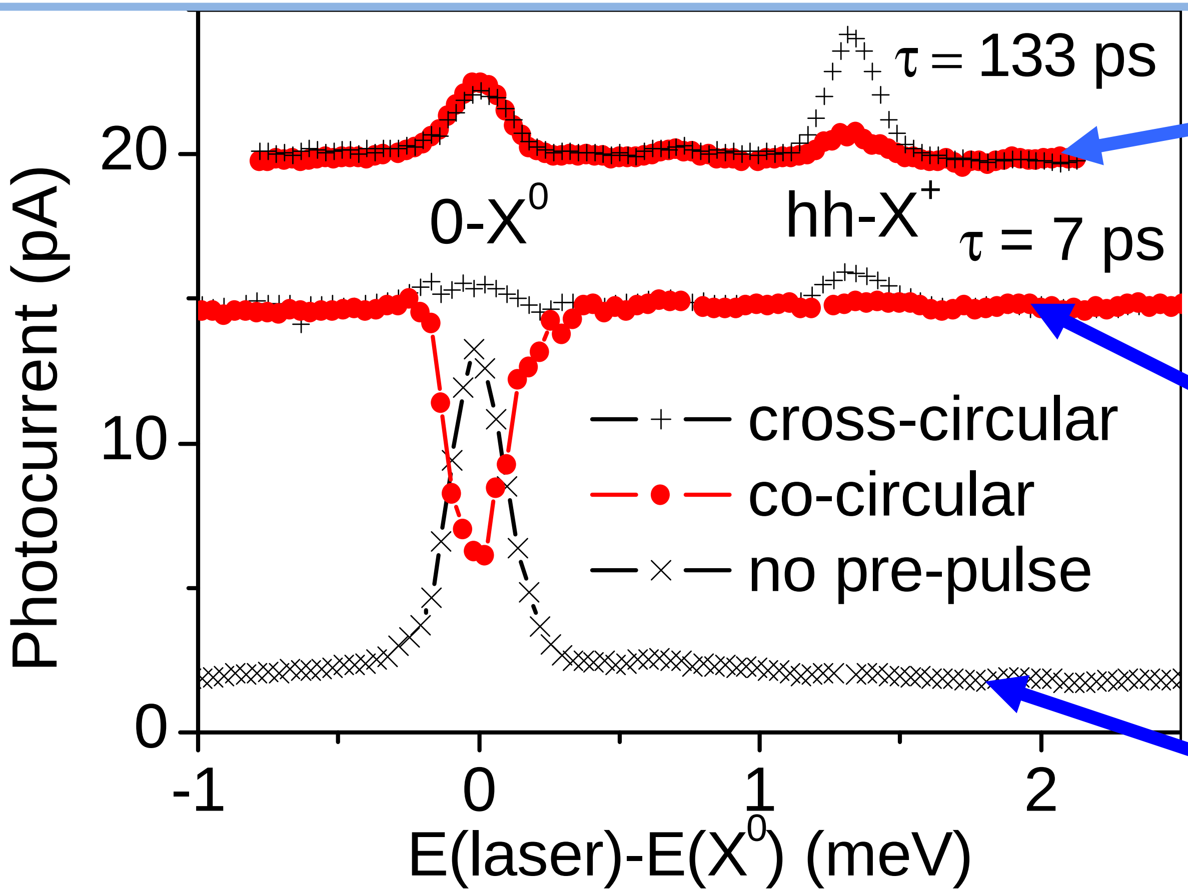




Example 3 cont: [Ramsay, PRL 100 097401 (2008). Two colour photocurrent spectrum, detection of single hole spin

Two pulses: "preparation" π-pulse (zero detuning) creates X^0

"Control" π-pulse variable detuning



Two pulse, time-delay>> electron tunnelling: sizeable polarization sensitive hh-X$^+$ peak observed

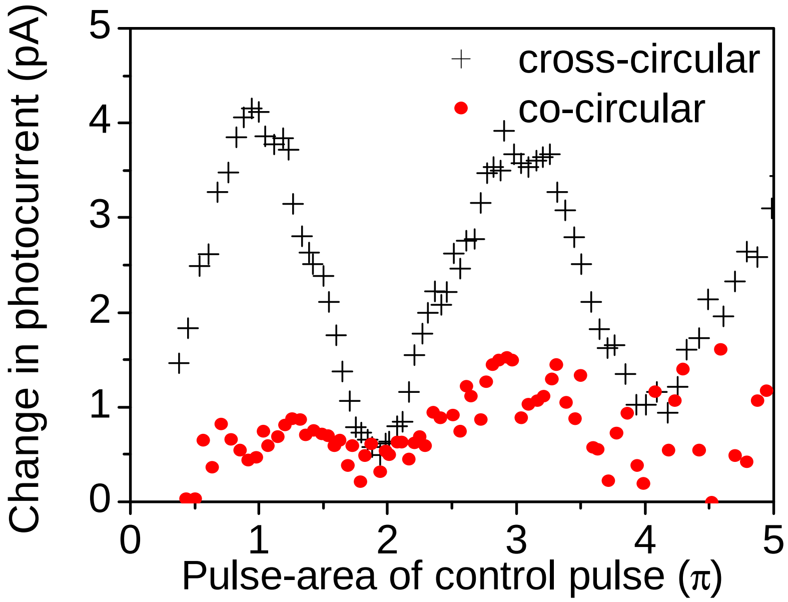
Two pulse, time-delay << electron tunnelling: Dip in 0-X^0 peak for co-circular excitation since pulse pair acts as 2π-pulse.

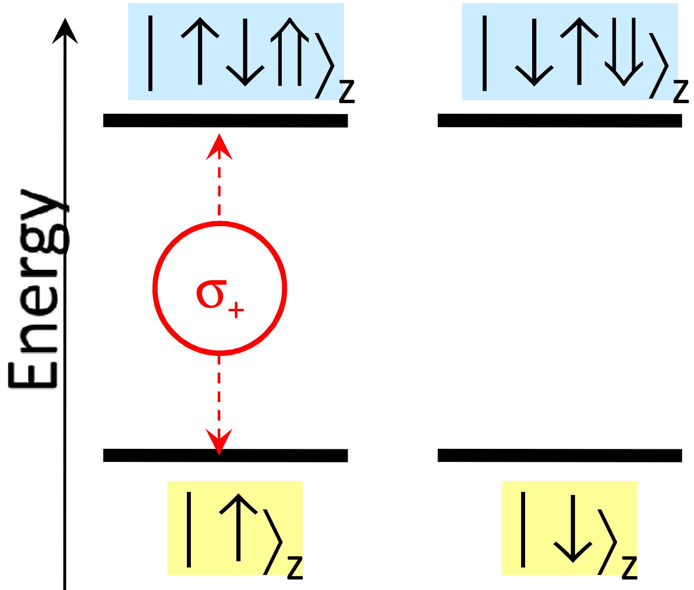
Single π-pulse:

Single peak 0-X^0

Example 3: [Ramsay, PRL 100 097401 (2008).] Rabi rotation conditional on initial hole spin state: single spin qubit phase-gate

Two pulses: "preparation" π-pulse (zero detuning) creates X0
"Control" π-pulse tuned to hh-X^{+} transition with variable pulse-area





Rabi rotation of positive trion transition
Conditional on spin, 2π-pulse gives phase-shift of π

Recall from 2-level atom case:
$\cos[\Theta(t)/2]|0;N\rangle - i.\sin[\Theta(t)/2]|X;N-1\rangle \rightarrow$
$-|0;N\rangle$, if $\Theta=2\pi$,
Geometric phase-shift
[Economou, PRL 99 207401 (2007).]

Part 4 summary

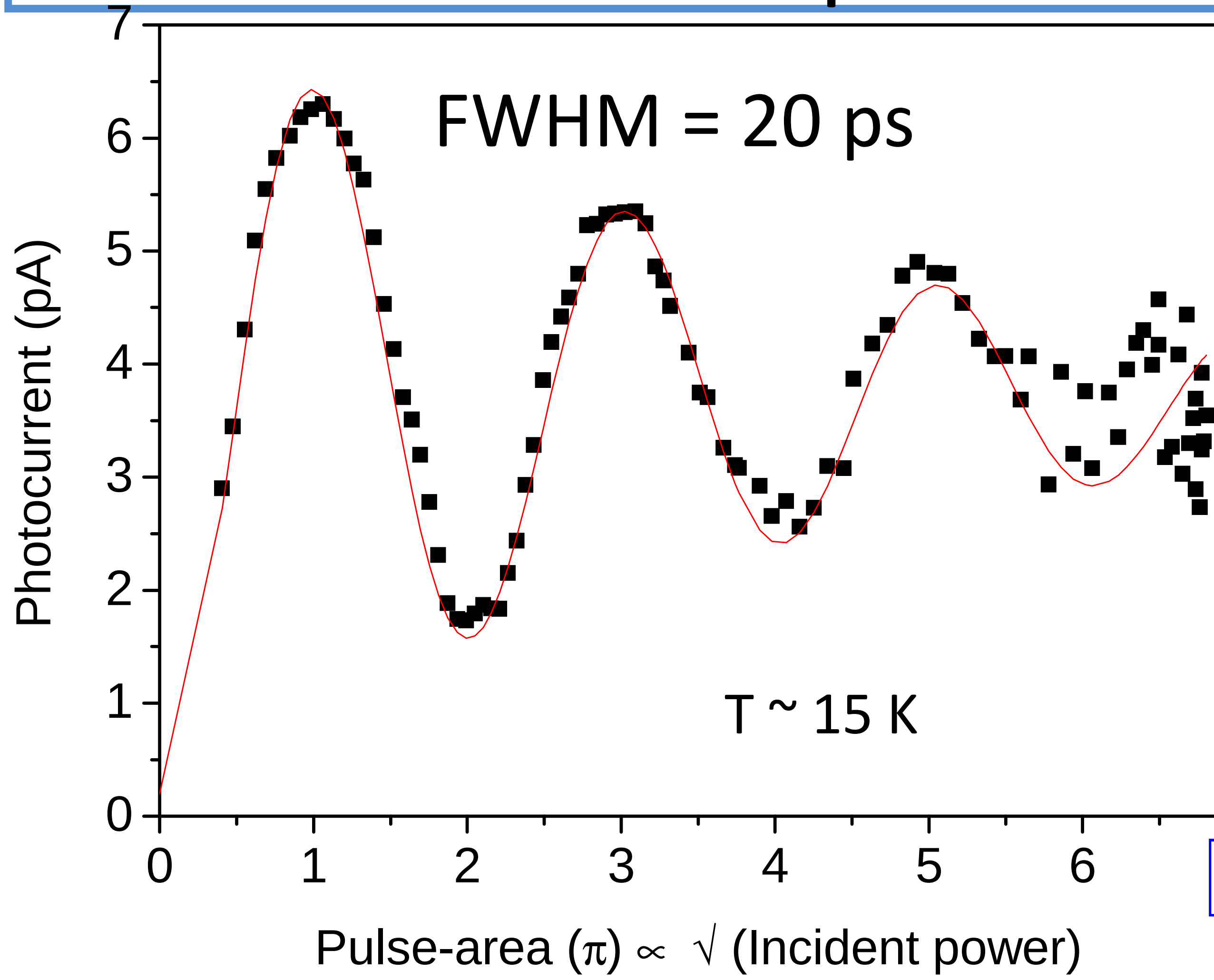
- Full coherent optical control of electron spin has been achieved [Press, Nature 418 232 (2008).]
- Partial optical control of hole spin has been achieved [Ramsay, PRL 100 097401 (2008).]
- Future work-
  - Double quantum dots
  - Try out other schemes for coherent control
  - Achieve coherent control in system with long coherence times
  - Achieve higher fidelity operations

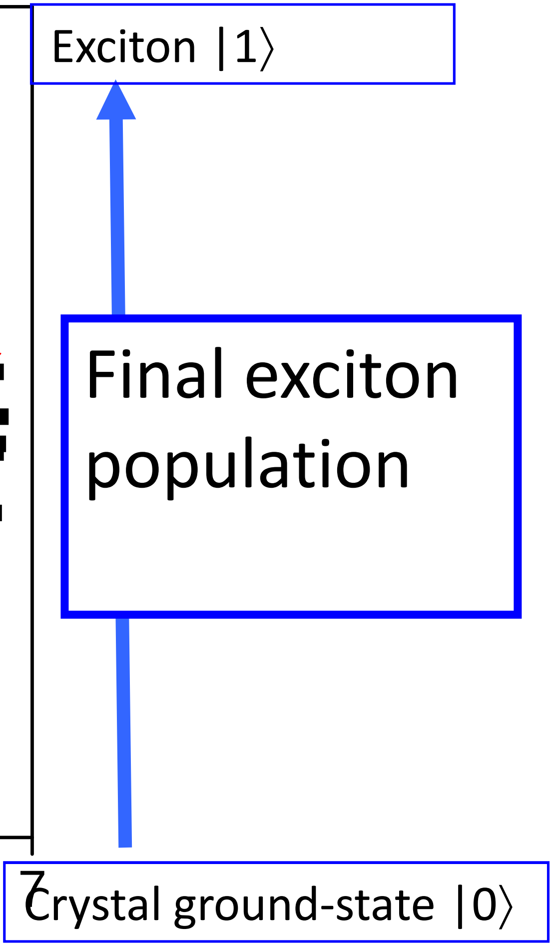
Part 5: Decoherence

- The quantum dot interacts with a solid-state environment
- Results in non-trivial dephasing dynamics
  - Acoustic phonons
  - Nuclear spin

Recent work identifying acoustic phonons as the main cause of the intensity damping observed in Rabi oscillations: Ramsay [ArXiv 0903.5278]

Why is the Rabi rotation intensity damped?



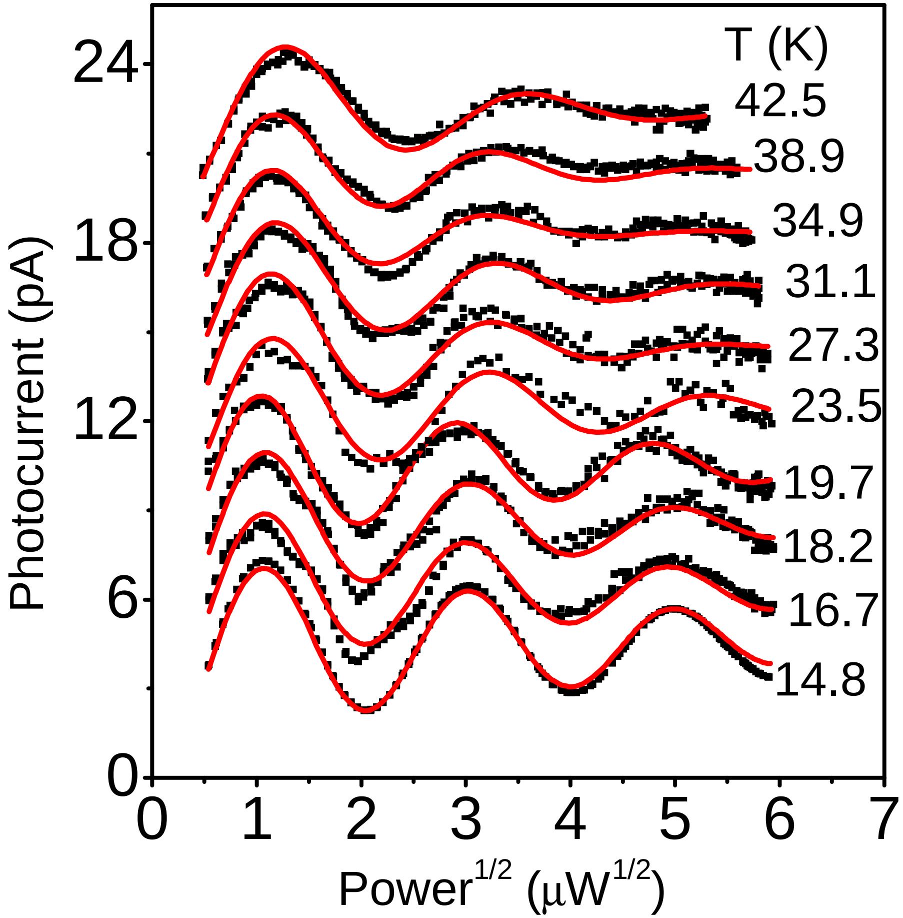


Exciton |1⟩

Final exciton population

Crystal ground-state |0⟩

Temperature dependence of Rabi rotations



Red fits to theory (later)

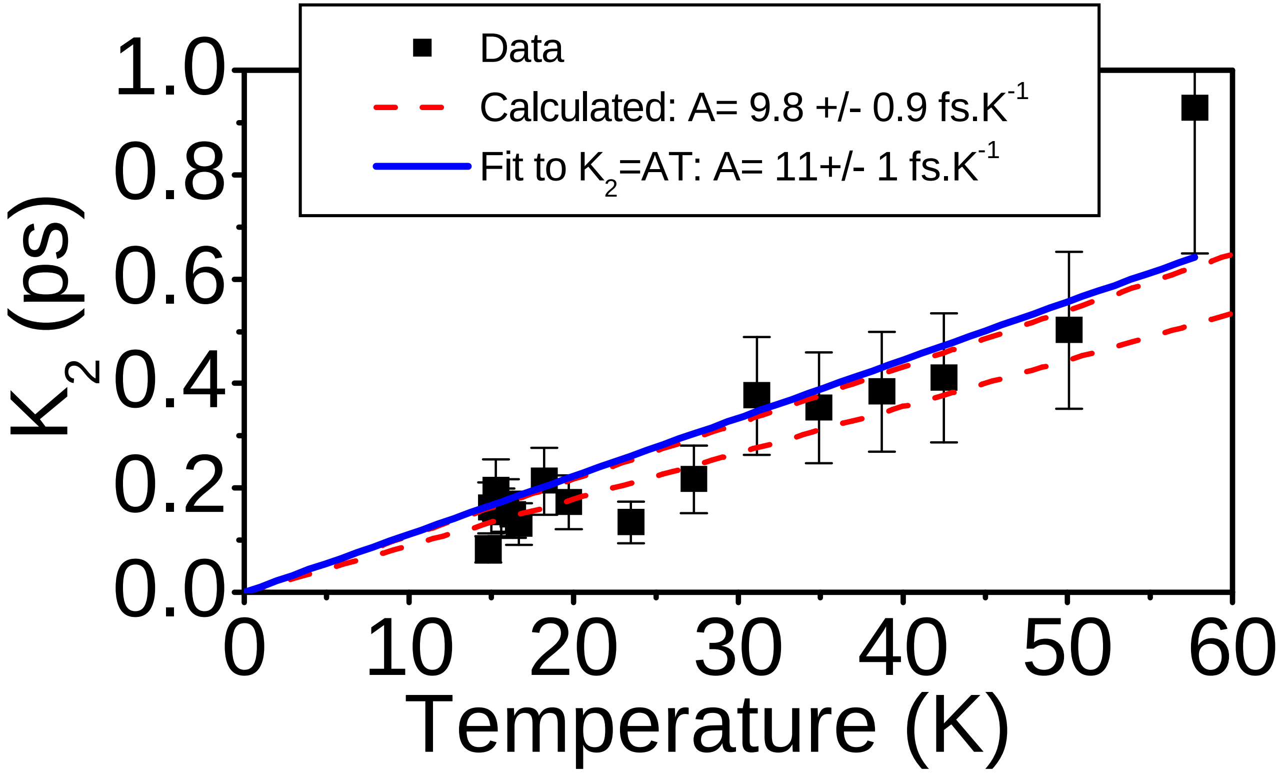
Damping increases with Temperature ⇒ phonons

Fit data to:

- $\partial_t \rho = \iota[\, H_c, \rho] + \partial_t \rho \,\big|_{\text{loss}}$
- $\partial_t \rho_{0X} \,\big|_{\text{loss}} = - K_2\, \Omega^2\, \rho_{0X}$

- Use fits of data to numerical solution of these equations to measure K_2
- Can calculate K_2 using acoustic phonon model, in Markov limit
- K_2 = AT, “A” depends on bulk material parameters only

Linear temperature dependence of excitation induced dephasing time $K_2 \Rightarrow$ Acoustic phonon model



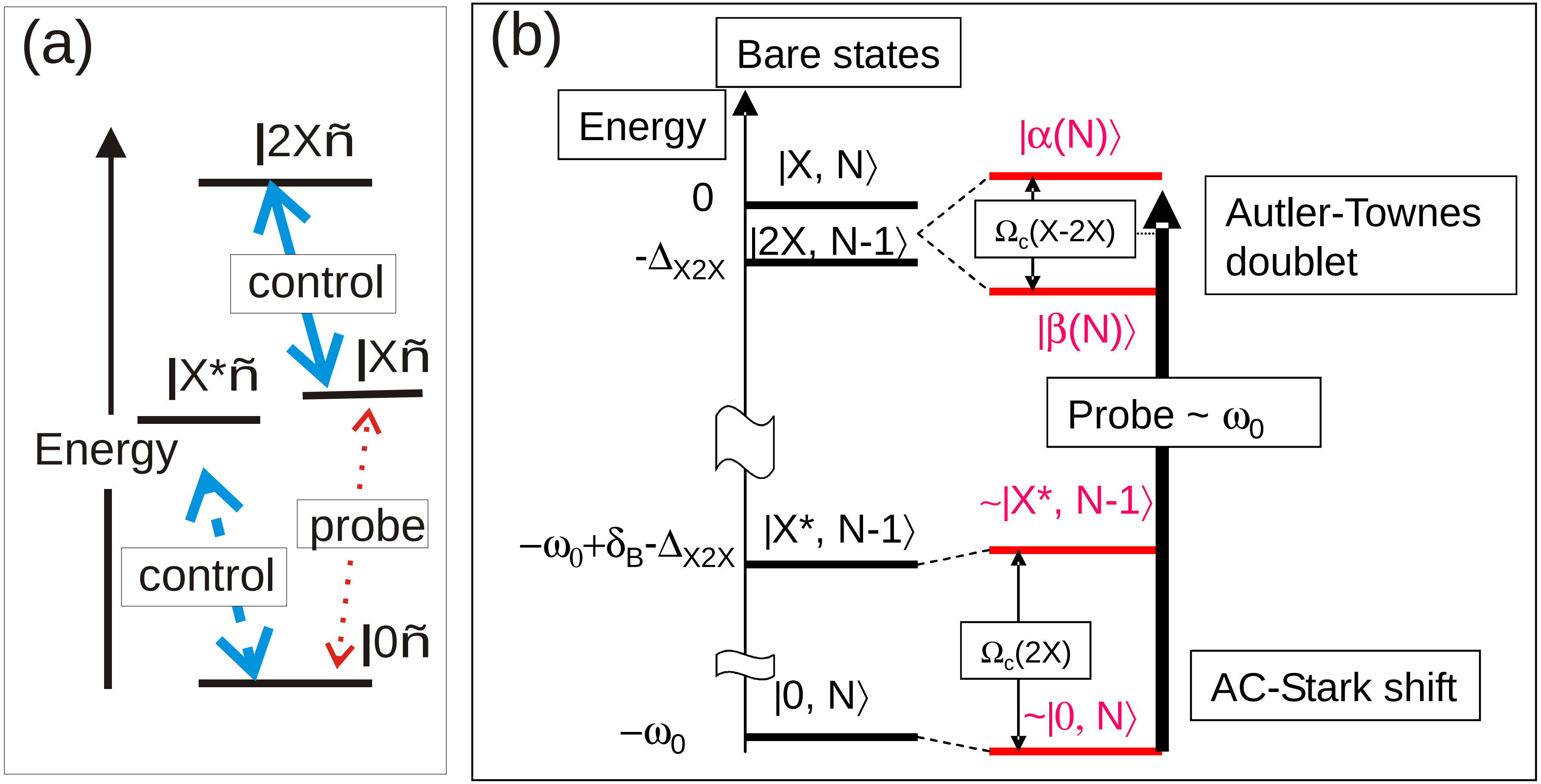
Use “A” to measure an effective deformation potential:

$|D_e - D_h|_{QD} = 9.0 \pm 0.3$ eV

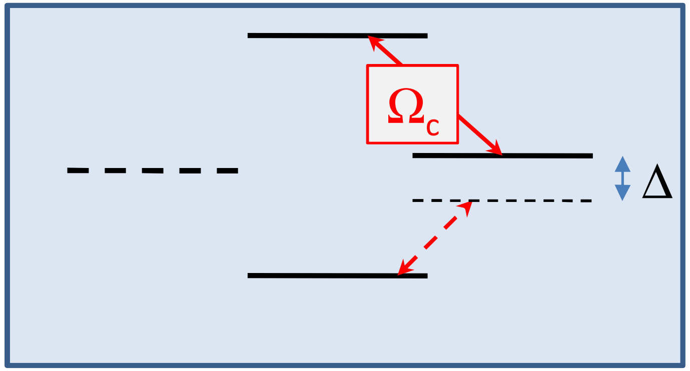
Part 5: Summary

- There is now experimental evidence to show that acoustic phonons do cause the intensity damping observed in Rabi oscillations

Energy-level diagram of 4-level exciton-biexciton system

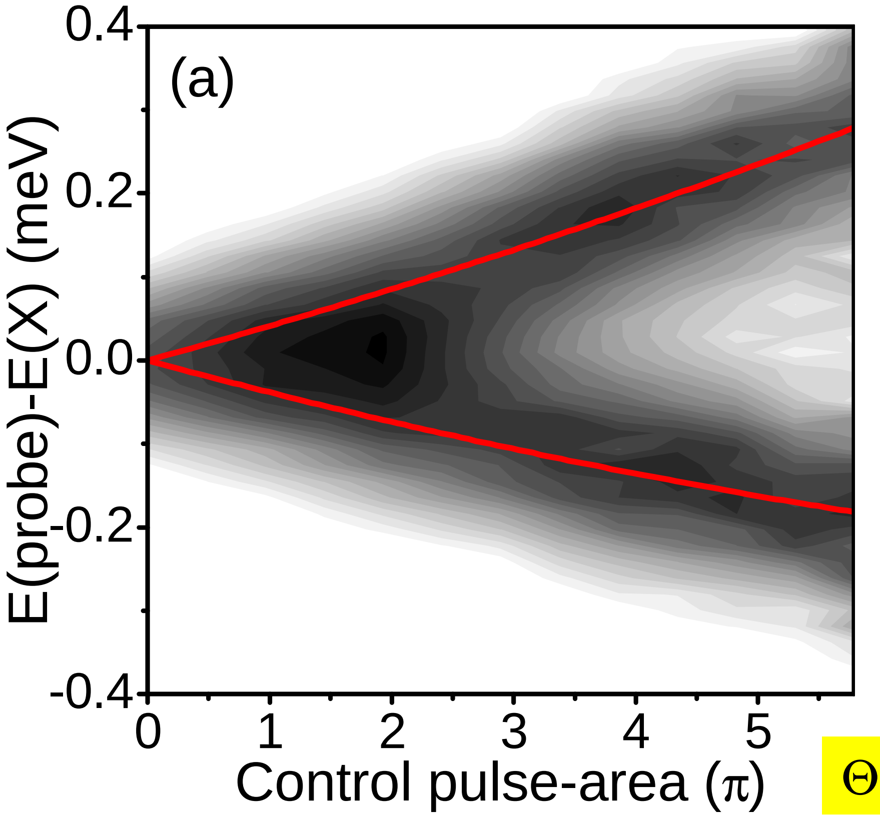


Power dependent Rabi-splitting

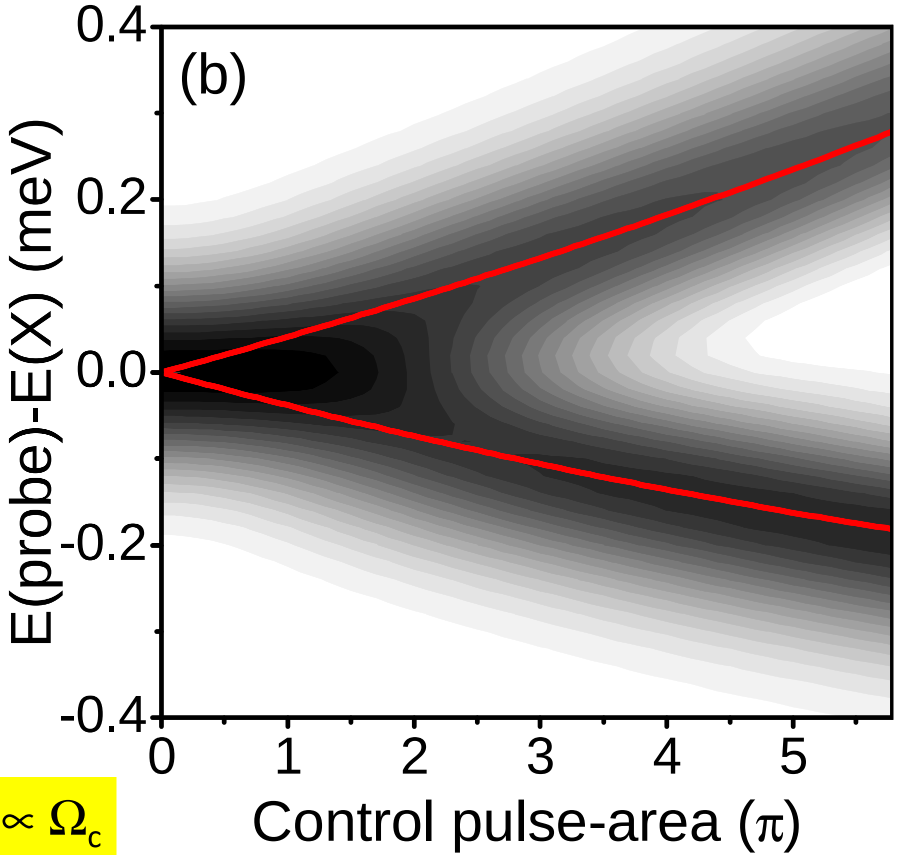


Δ

Experiment

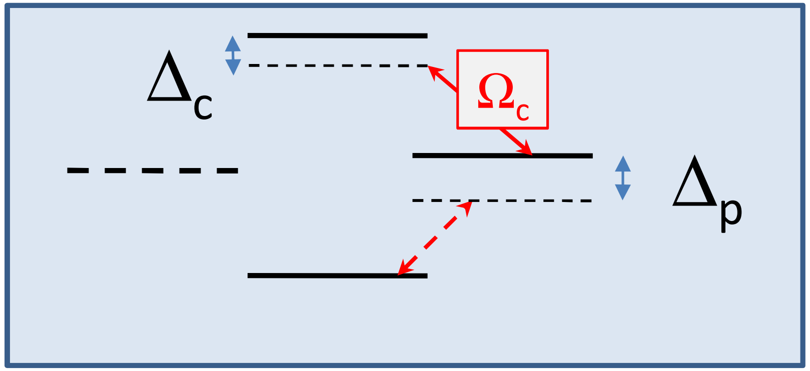


Theory



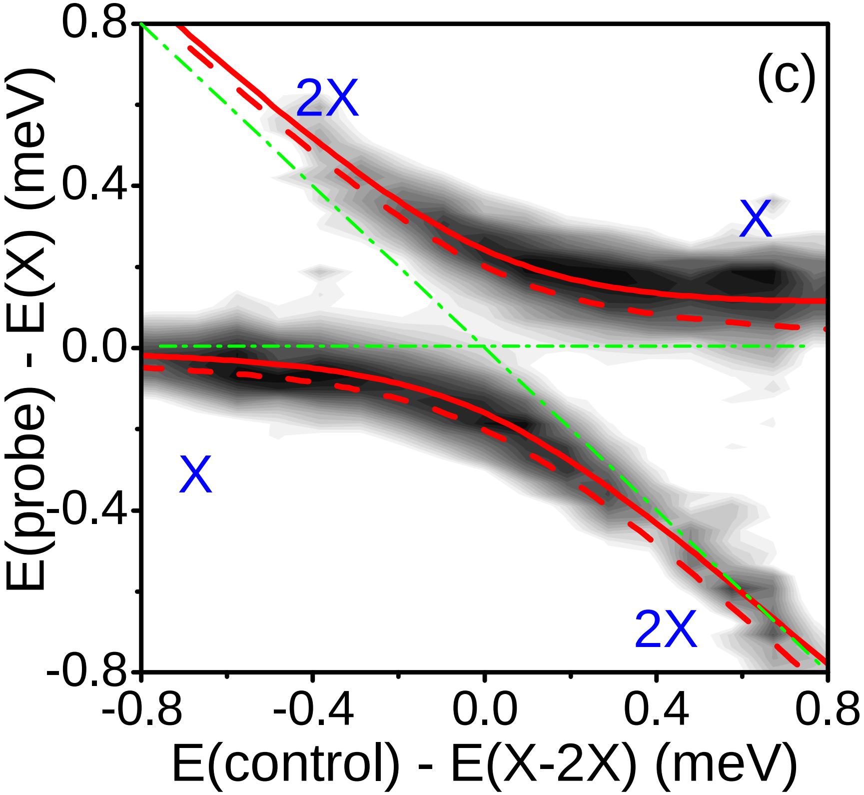
$\Theta \propto \Omega_c$

Anti-crossing of dressed states

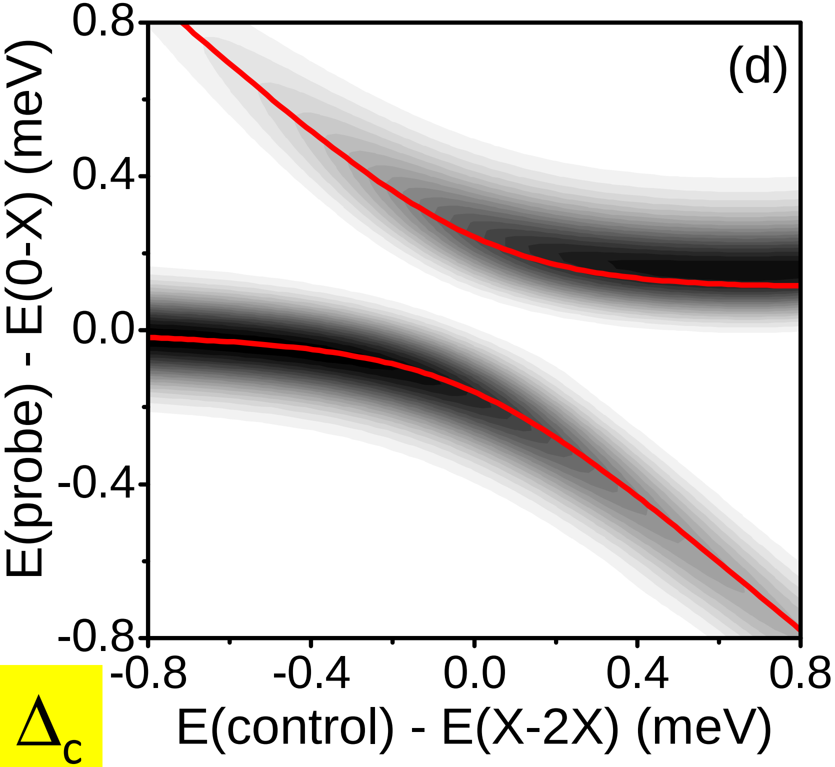


Experiment

Δ_p

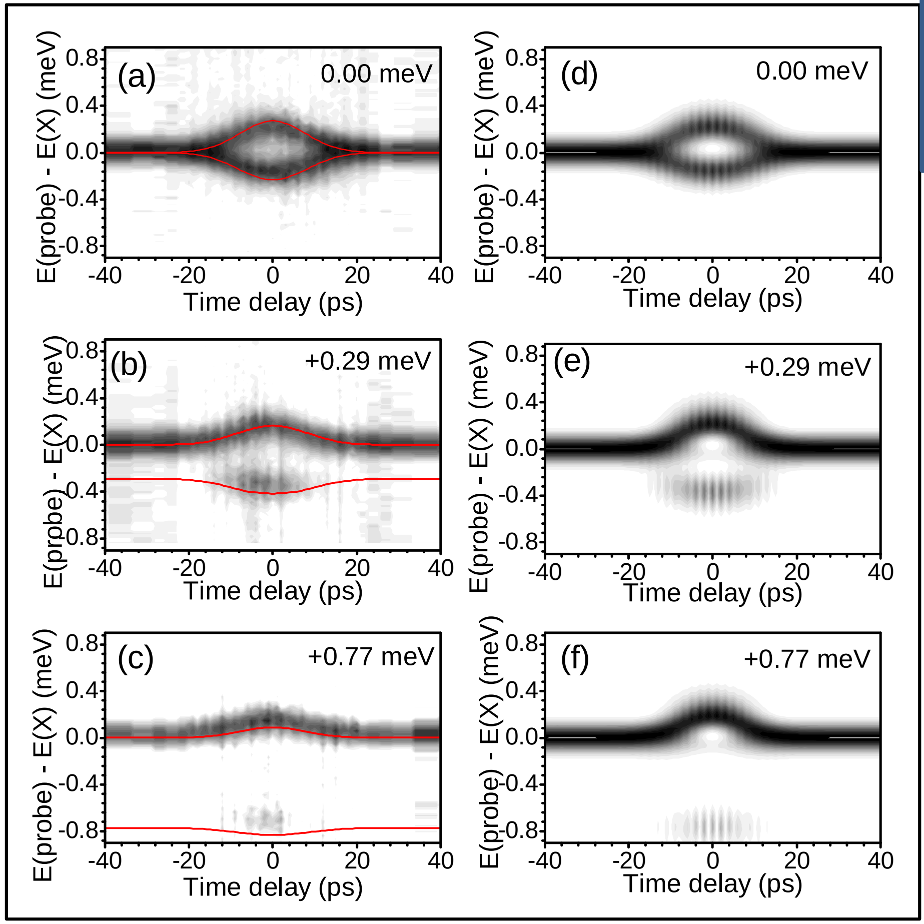


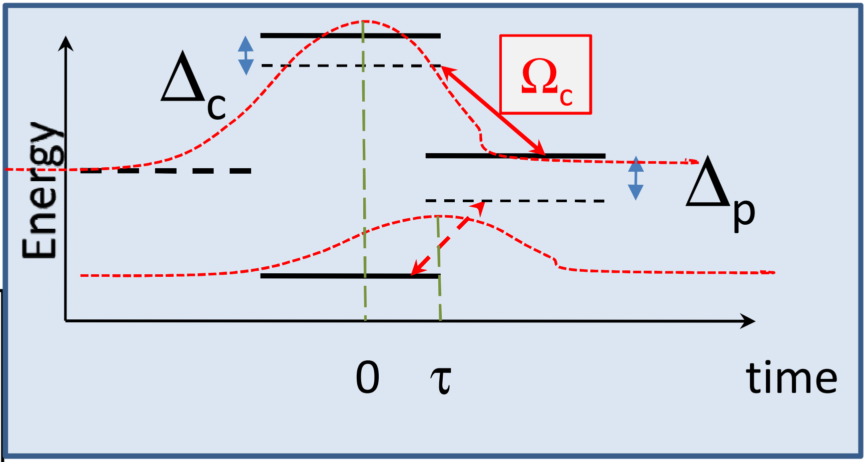
Theory



Δ_c

Time evolution of splitting





Splitting follows the control pulse

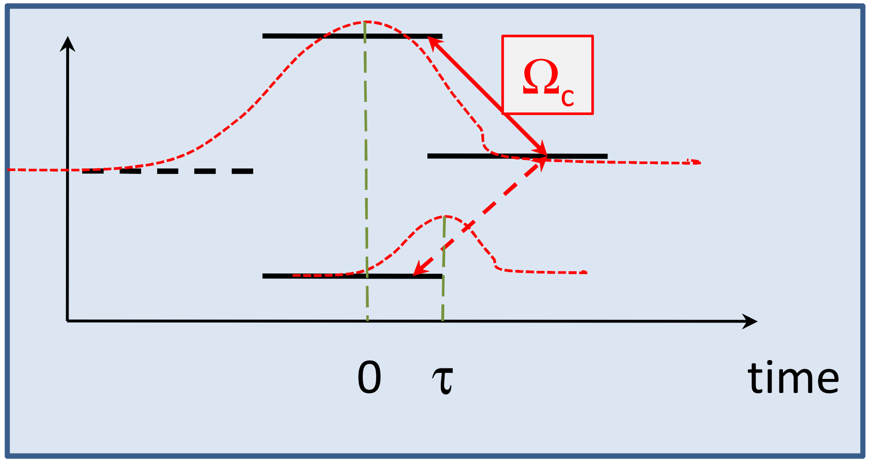
⇒ Coherent effect (i.e. no field, no splitting)

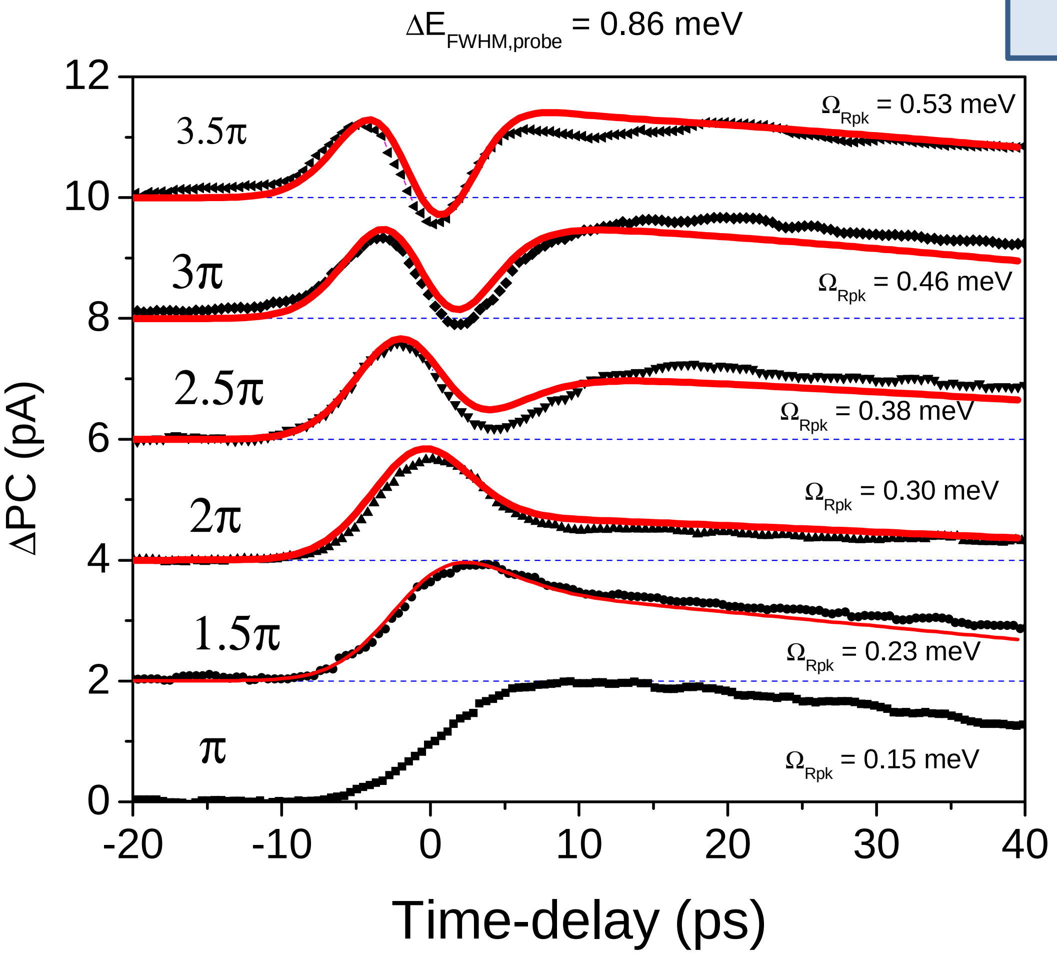
⇒ Not a result of real carrier nonlinearity

⇒ We can manipulate the Rabi splitting, and the admixture of the dressed states on picosecond timescales.

⇒ Demonstrates the feasibility of using adiabatic passage techniques for control of excitons or spins [e.g. Calarco PRA 2003]

Time-resolved Rabi oscillation of X-2X transition





First observation of Rabi oscillation in time domain, at fixed incident power for QD

Can fit the data by assuming an intensity dependent dephasing term

Summary

- An intense laser pulse couples the exciton and biexciton states to form an Autler-Townes doublet
- We observe the doublet in both time and frequency domains
- Also observe AC-Stark effect
- We show that the Rabi-splitting, and hence the composition of the dressed states follow the control field, demonstrating the feasibility of using adiabatic passage techniques for the control of excitons and spins
- We observe beat between X/2X dressed states, time-resolving the X-2X Rabi oscillation

Selected publications on coherent control of quantum dots

1. **Excitation induced dephasing of quantum dot excitonic Rabi rotations,** *A. J. Ramsay, Achanta Venu Gopal, E. M. Gauger, A, Nazir, B. W. Lovett, A. M. Fox, and M. S. Skolnick,* ArXiv: 0903.5278
2. **Beating of exciton dressed states in single InGaAs/GaAs quantum dot***,*
   S. J. Boyle, A. J. Ramsay, A. P. Heberle, M. Hopkinson, A. M. Fox, and M. S. Skolnick, Physical Review Letters **102** 097401 (2009).
3. **Two-qubit conditional quantum-logic operation in a single self-assembled quantum dot**
   S. J. Boyle, A. J. Ramsay, F. Bello, H. Y. Liu, M. Hopkinson, A. M. Fox, and M. S. Skolnick
   Physical Review B **78** 075301 (2008)
4. **Fast Optical Preparation, Control, and Readout of a Single Quantum Dot Spin**
   A. J. Ramsay, S. J. Boyle, R. S. Kolodka, J. B. B. Oliviera, J. Skiba-Szymanska, H. Y. Liu, M. Hopkinson, A. M. Fox, and M. Skolnick
   Physical Review Letters **100** 197401 (2008)
5. **Inversion recovery of single quantum-dot exciton based qubit**
   R. S. Kolodka, A. J. Ramsay, J. Skiba-Szymanska, P. W. Fry, H. Y. Liu, A. M. Fox and M. S. Skolnick
   Physical Review B **75** 193306 (2007)
6. **Coherent response of a quantum dot exciton driven by a rectangular spectrum optical pulse**
   A. J. Ramsay, R. S. Kolodka, F. Bello, P. W. Fry, W. K. Ng, A. Tahraoui, H. Y. Liu, M. Hopkinson, D. M. Whittaker, A. M. Fox, and M. S. Skolnick
   Physical Review B **75** 113302 (2007)