

# Coherent control and electrical detection of charge excitations in hydrogenic silicon impurities with a Free Electron Laser

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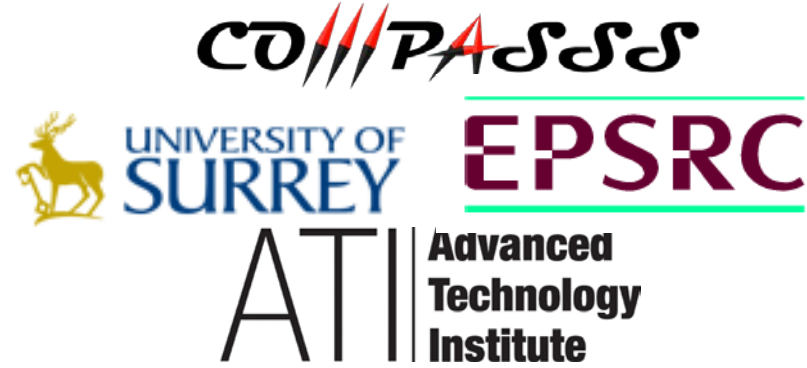
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Coherent Optical and Microwave Physics for Atomic Scale Spintronics in Silicon



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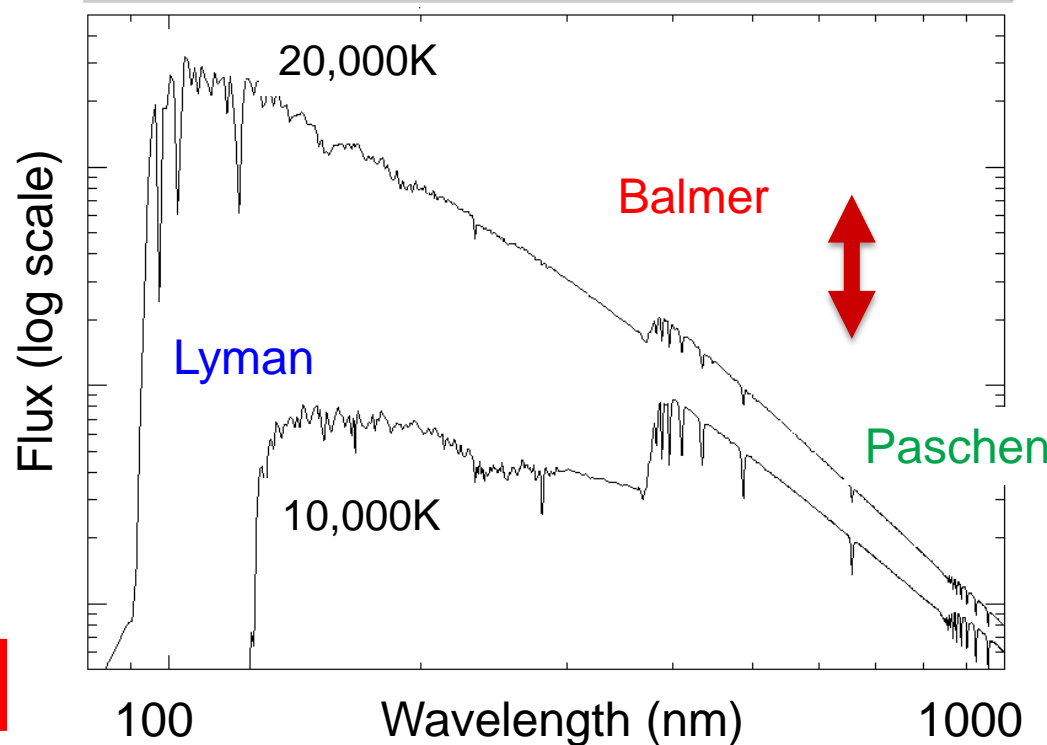
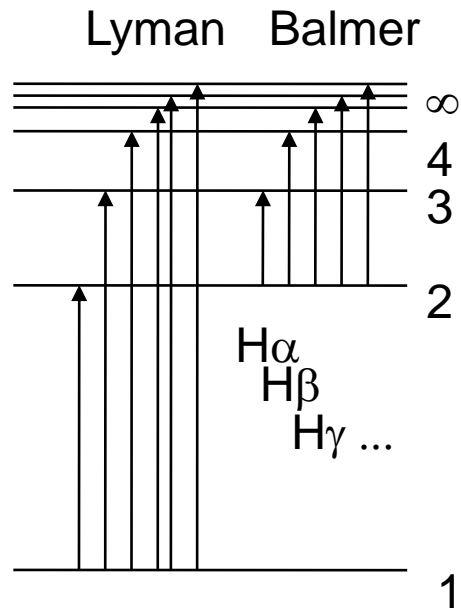
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# Rydberg spectrum of hydrogen

- Hydrogen absorption spectrum seen superimposed on the emission from a very hot black body (a star)

$$\frac{1}{\lambda_{nm}} = R \left[ \frac{1}{n^2} - \frac{1}{m^2} \right]$$

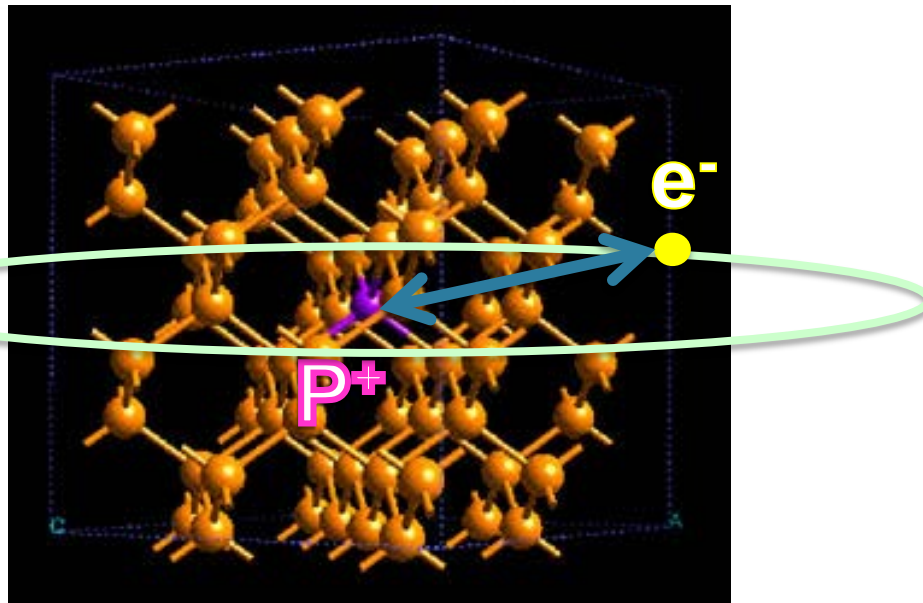


$H\alpha$  line:  $E_3 - E_2 = \frac{5}{36} 13.6\text{eV} \equiv 656\text{nm}$

# A hydrogen-like atom in a silicon chip: The Group 5

impurity

1	2											3	4	5	6	7	0
		H															He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg							



- P looks like silicon with
  - an extra +ve charge in the ion
  - an extra electron orbiting
- The electron-ion attraction is screened by  $\epsilon_r$
- The mass is reduced by  $m^*$

# Scaling from hydrogen to donor

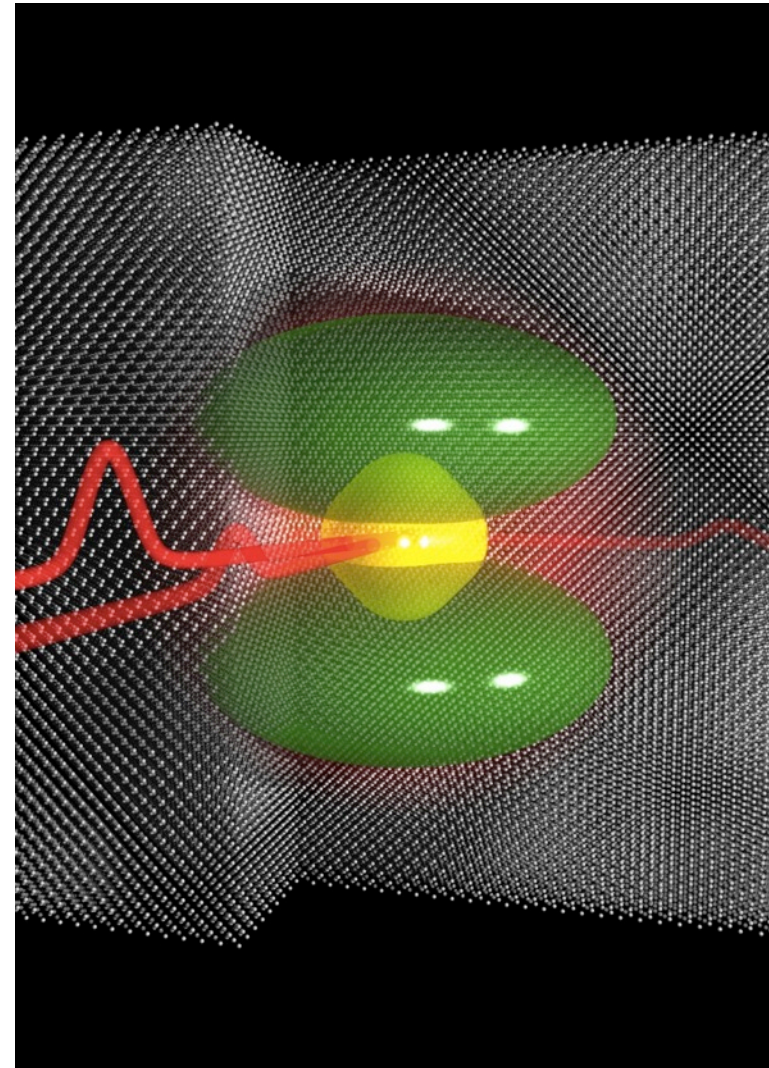
- Binding energy:

$$E_R = \frac{1}{2} \left( \frac{e^2}{4\pi\hbar} \right)^2 \frac{m_e}{\epsilon^2}$$

- Bohr radius

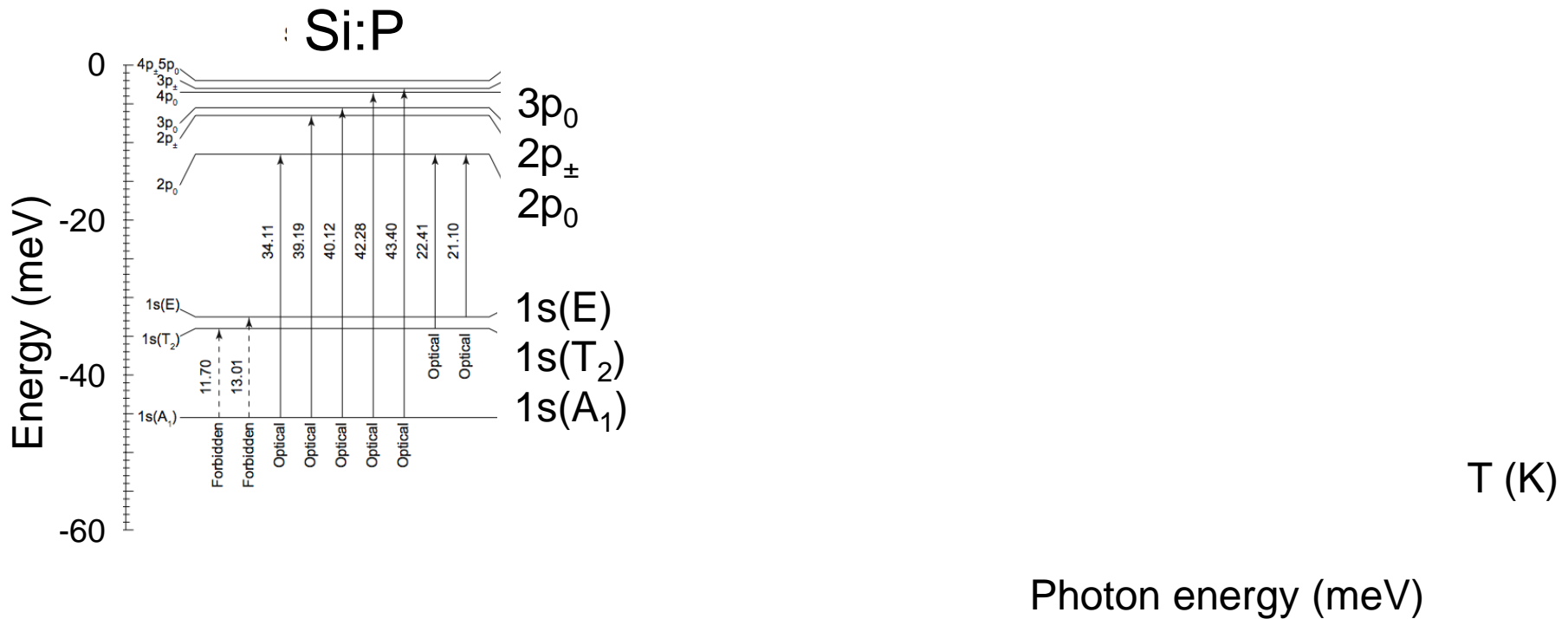
$$a_0 = \frac{4\pi\hbar^2}{e^2} \frac{\epsilon}{m_e}$$

	H	Si:P
$\epsilon_r$	1	11.4
$m_e$	1	0.19
$E_R$	13.6 eV	0.020 eV
$a_0$	0.056 nm	3.2 nm



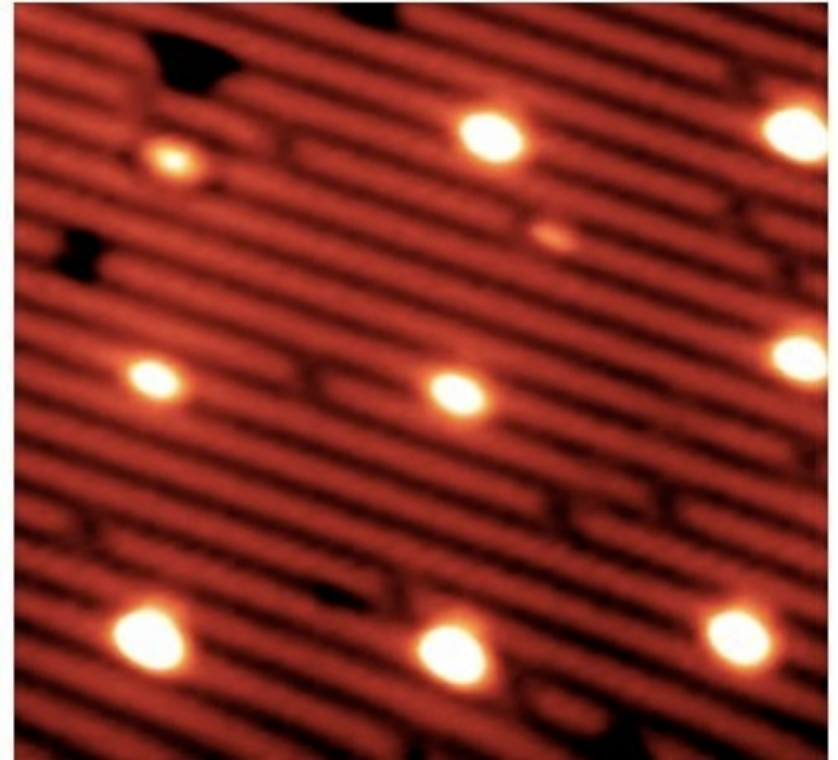
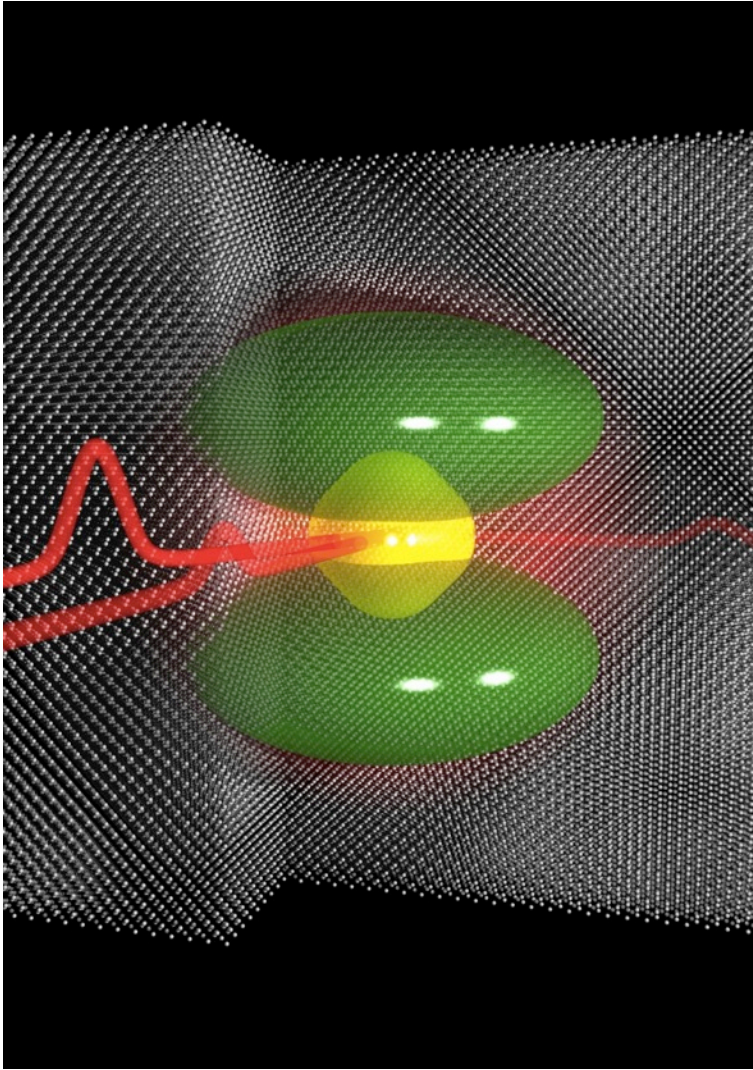
# Hydrogen like spectrum in Si:P

SA Lynch et al, Phys. Rev. B (2010)



- Some qualitative differences between hydrogen and donor
  - States split by cubic crystal symmetry (e.g. 2p<sub>0</sub> and 2p<sub>±</sub>)
  - s-states shifted down in energy by “quantum defect” (a.k.a. “chemical shift” or “central cell correction”) due to the P<sup>+</sup> ion not being point-like

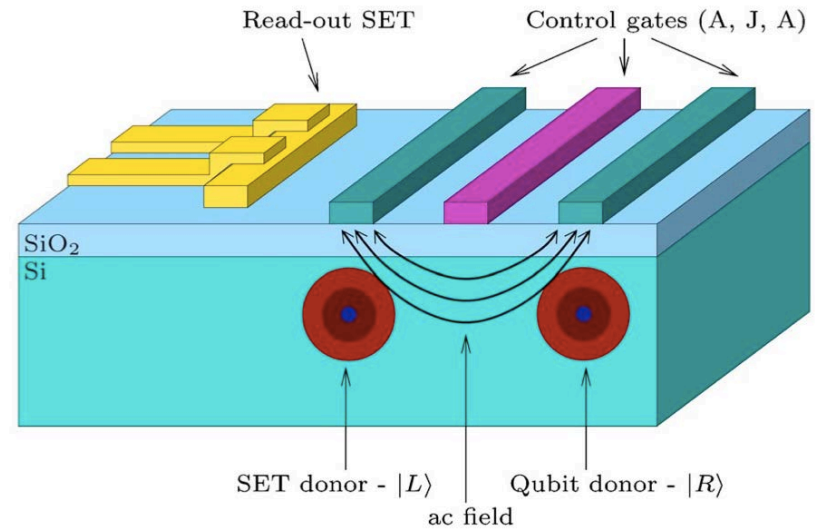
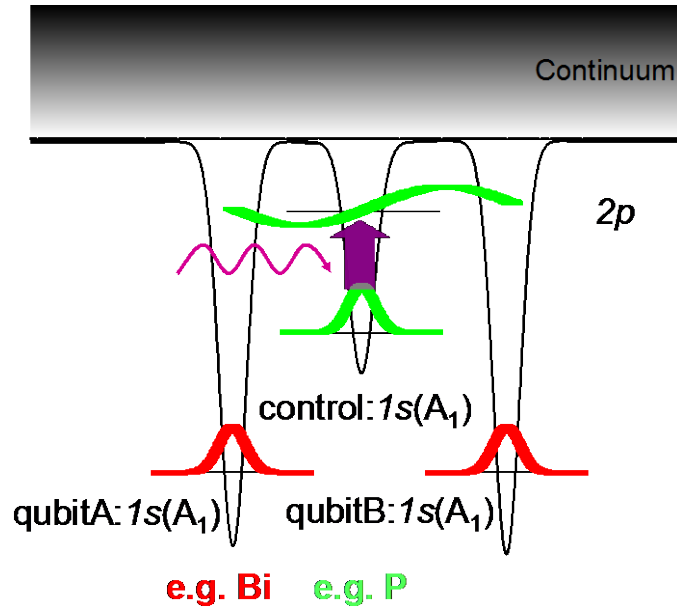
# Control of donors in time & space





# Silicon atomic scale electronics

- a) Silicon is easy to process into sophisticated electronic devices  $\Rightarrow$  good connection to impurity
- b) Impurities are exactly reproducible  $\Rightarrow$  scalable
- c) Silicon donors can be positioned with single atom precision  $\Rightarrow$  large registers
- d) Si:P electron spin  $T_2$  up to millisecc and nuclear spin  $T_2$  is 3 hours!  $\Rightarrow$  good quantum memory
- e) Si:P orbitronics (our work): Rydberg excitations with wavefunction radius up to 100nm (!!!) are lifetime broadened with  $T_2$  up to 0.1ns  $\Rightarrow$  good electric dipolar quantum control



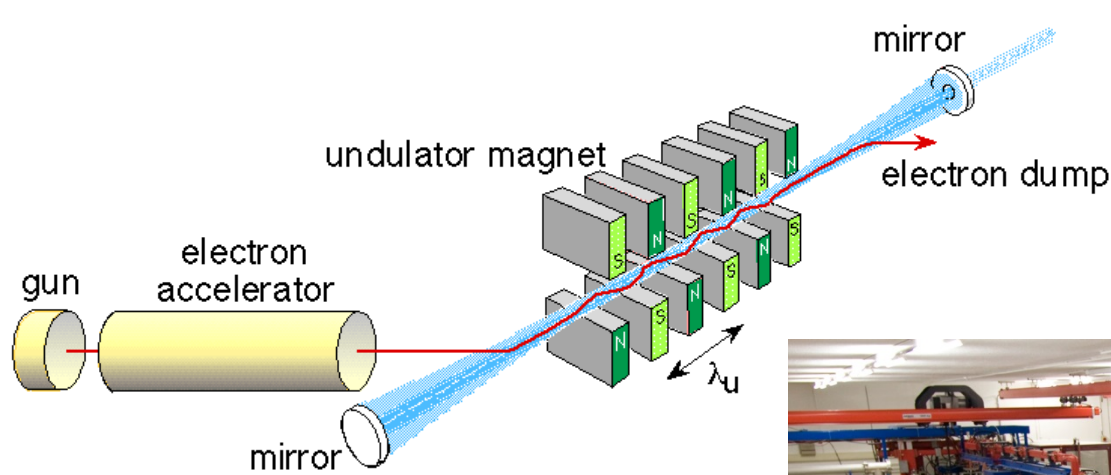
- Stoneham-Fisher-Greenland scheme: THz gated entanglement/control/gating between qubits

A. M. Stoneham *et al*, *J. Phys. C*, 15, L447, 2003.

- Kane/Hollenberg scheme: THz induced spin-to-charge conversion between qubit and SET donors

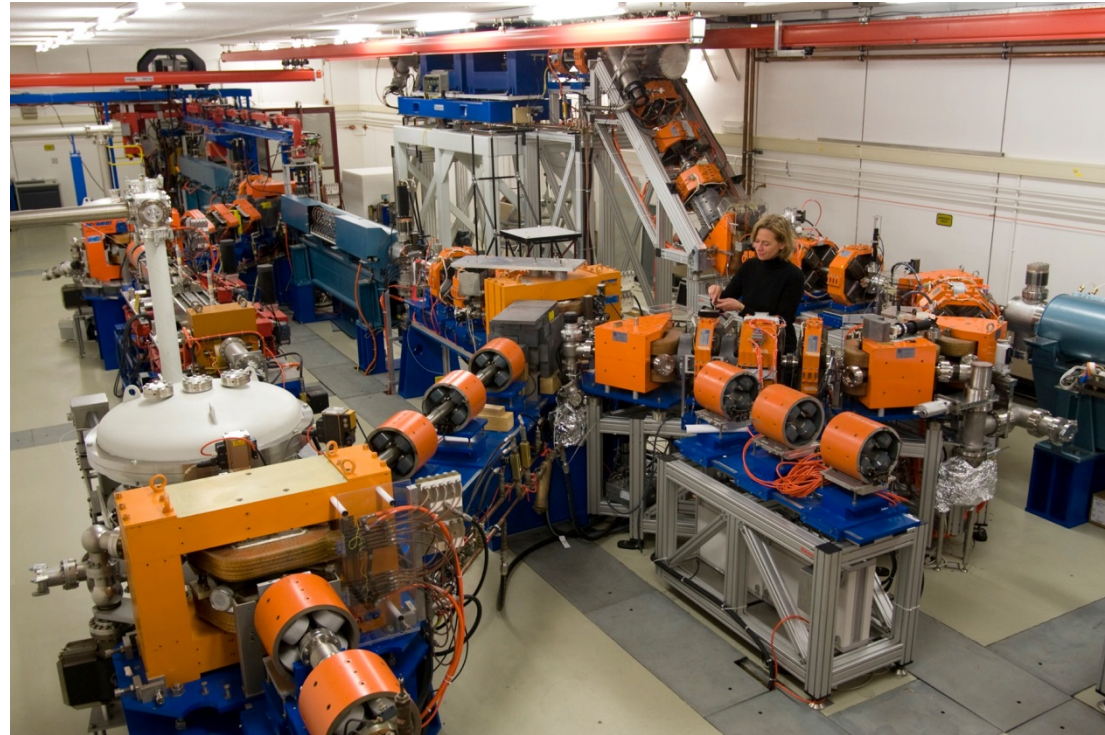
L.C.L.Hollenberg *et al* *Phys.Rev.B* 69, 233301 (2004)

# Importance of THz in Si QIP



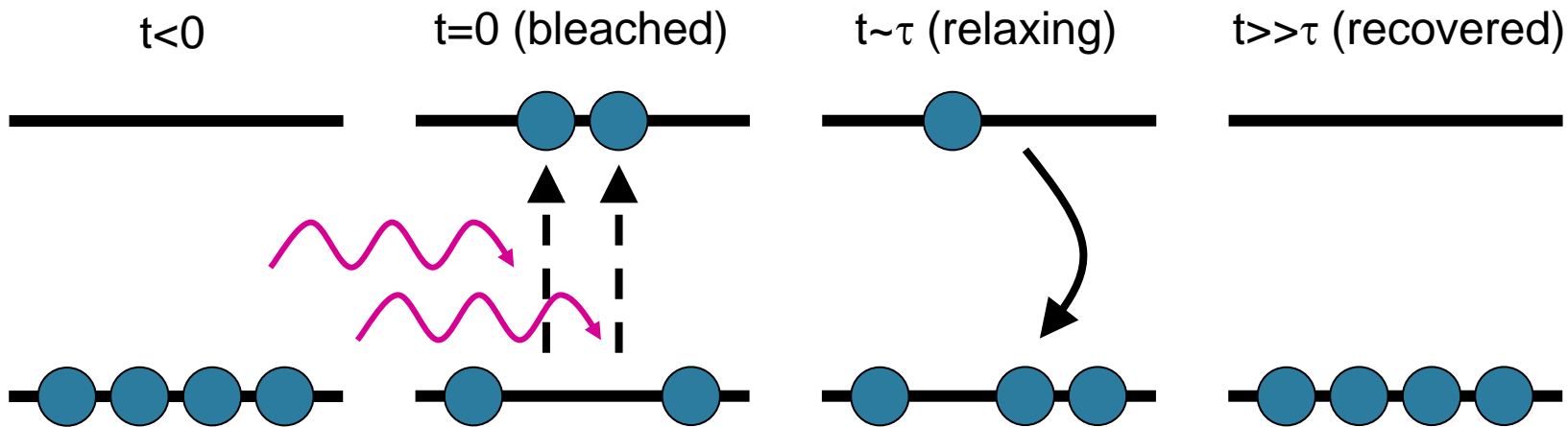
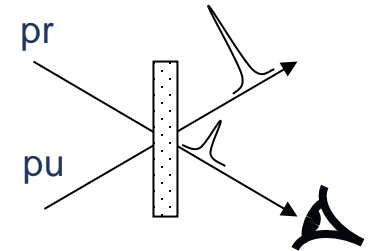
$$n \lambda_s = \frac{\lambda_u (1 + K^2)}{2 \gamma^2}$$

- wavelength
  - 4-250μm
  - 1-100THz
- Pulse duration
  - 1-10 ps
- Pulse energy
  - ~10μJ

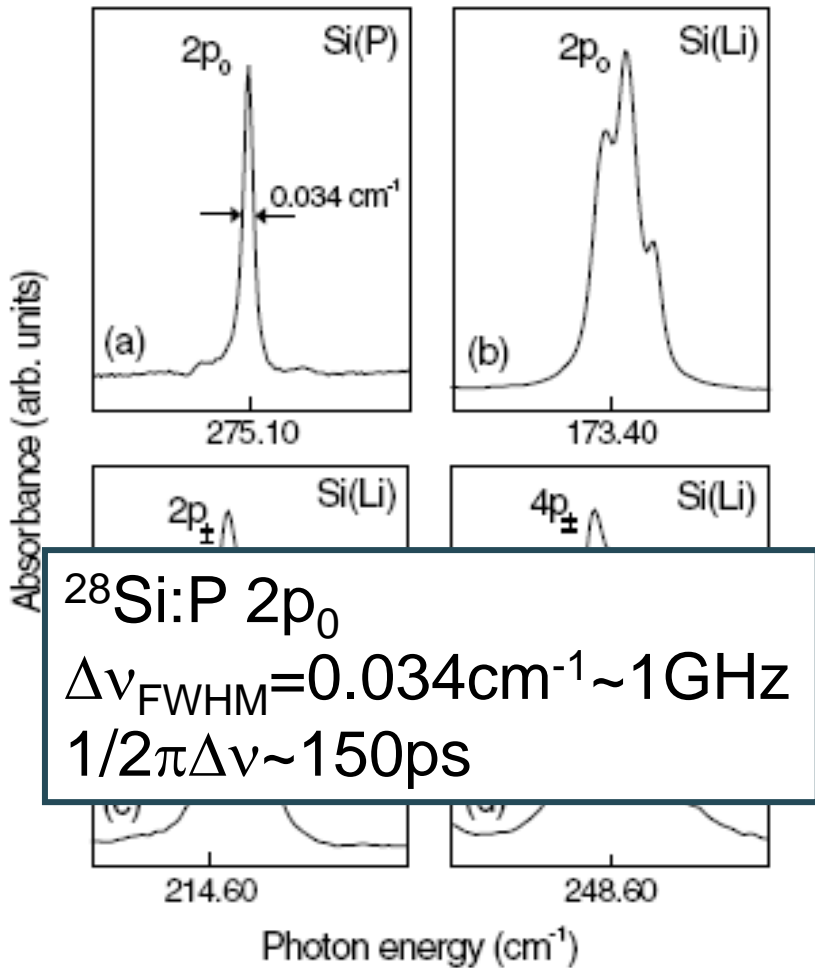


# FELIX, the Free-Electron Laser for Infrared eXperiments (Nijmegen)

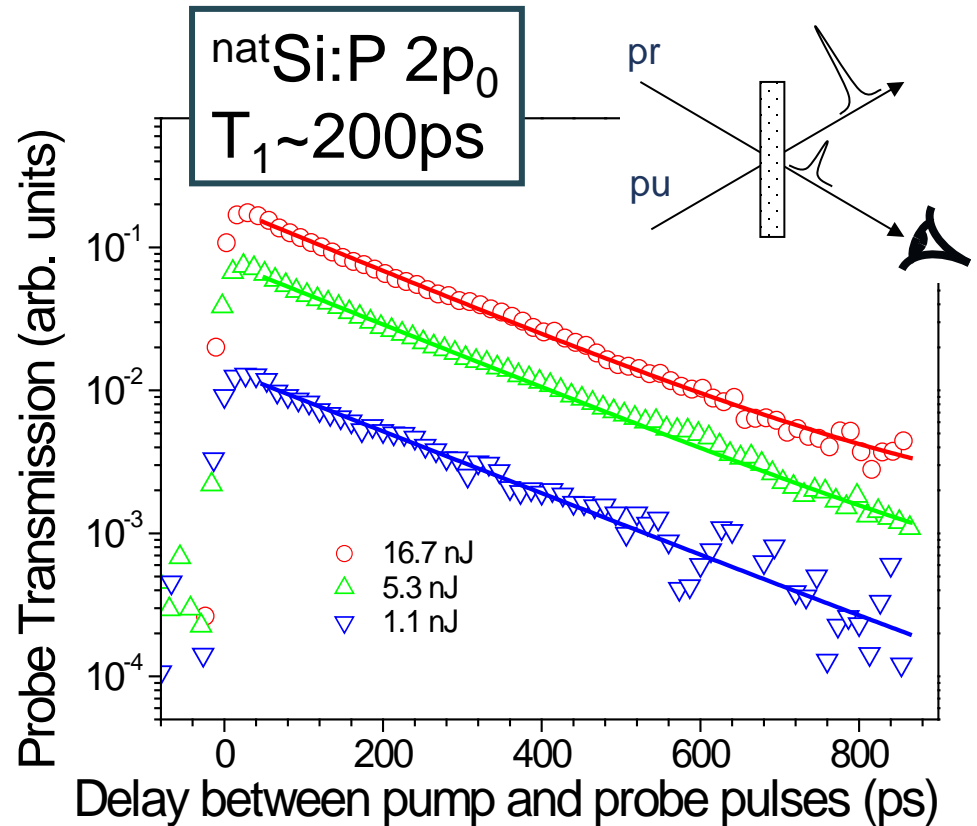
- A strong pump pulse bleaches the transmission by exciting  $\sim 50\%$  of the oscillators
- A weak probe measures the transmission recover as a function of time delay



# Incoherent dynamics experiment: pump-probe



Thewalt *et al.* *PRL* **90** (2003)



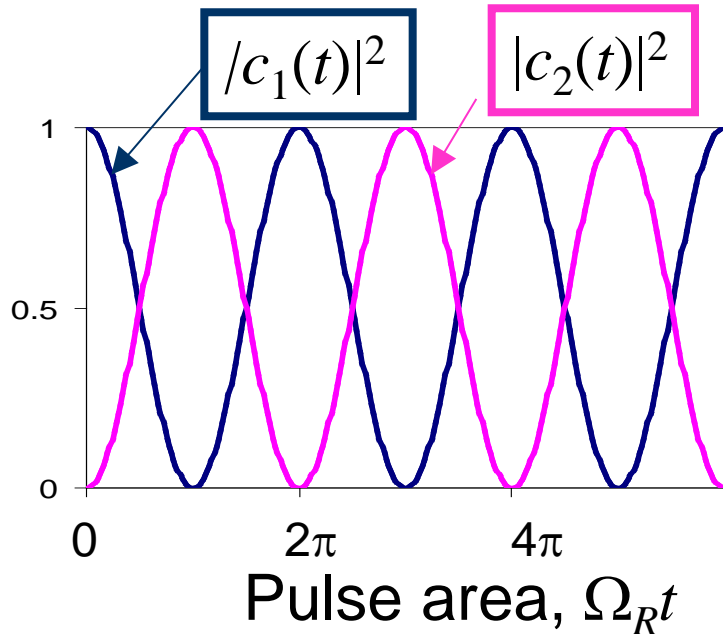
NQ Vinh *et al.* *PNAS* **105**, 10649 (2008)

NQ Vinh *et al.* *PRX* **3**, 3, 011019 (2013)

HW Hubers *et al.* *PRB* **88**, 035201 (2013)

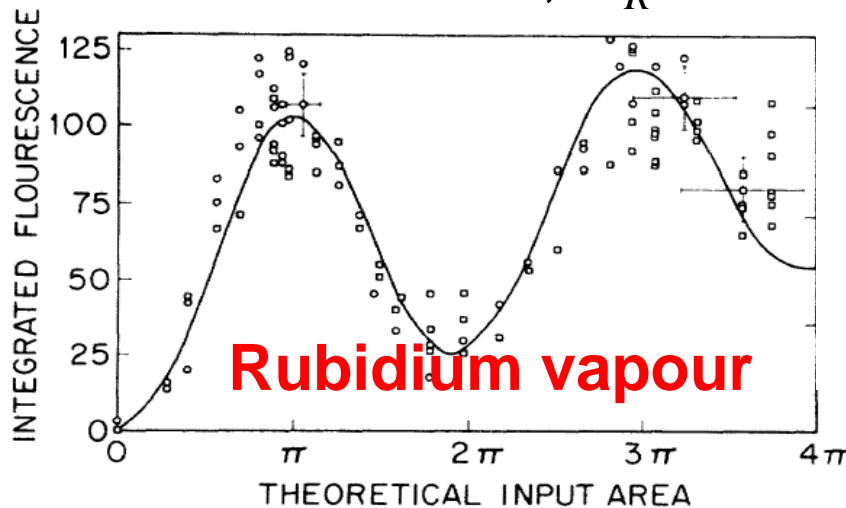
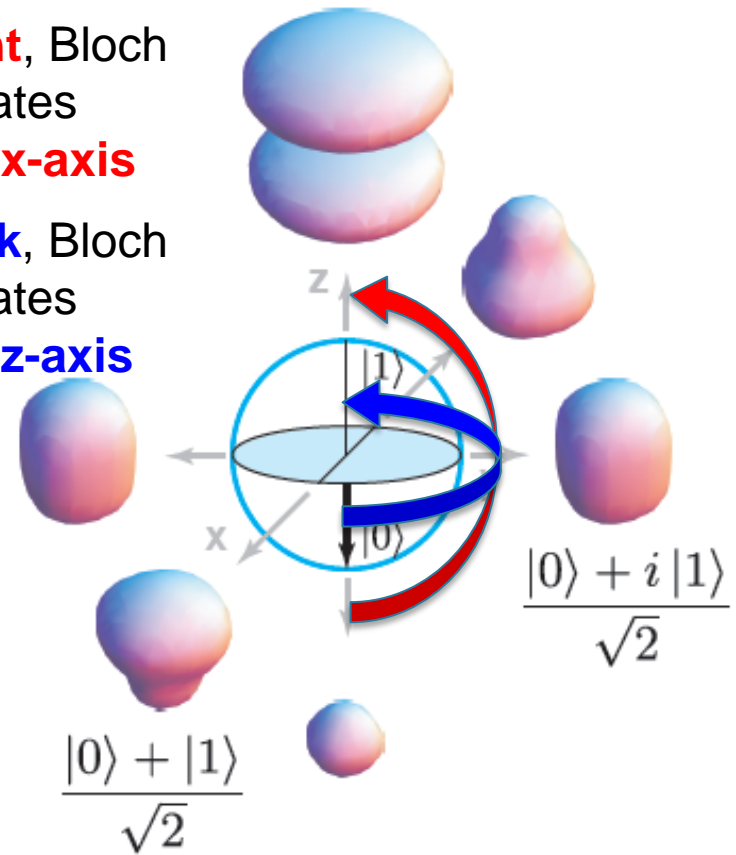
# Spectral and time domain spectroscopy of orbital excitations

# Dynamics during light pulse: Rabi flopping



In the **light**, Bloch vector rotates about the **x-axis**

In the **dark**, Bloch vector rotates about the **z-axis**

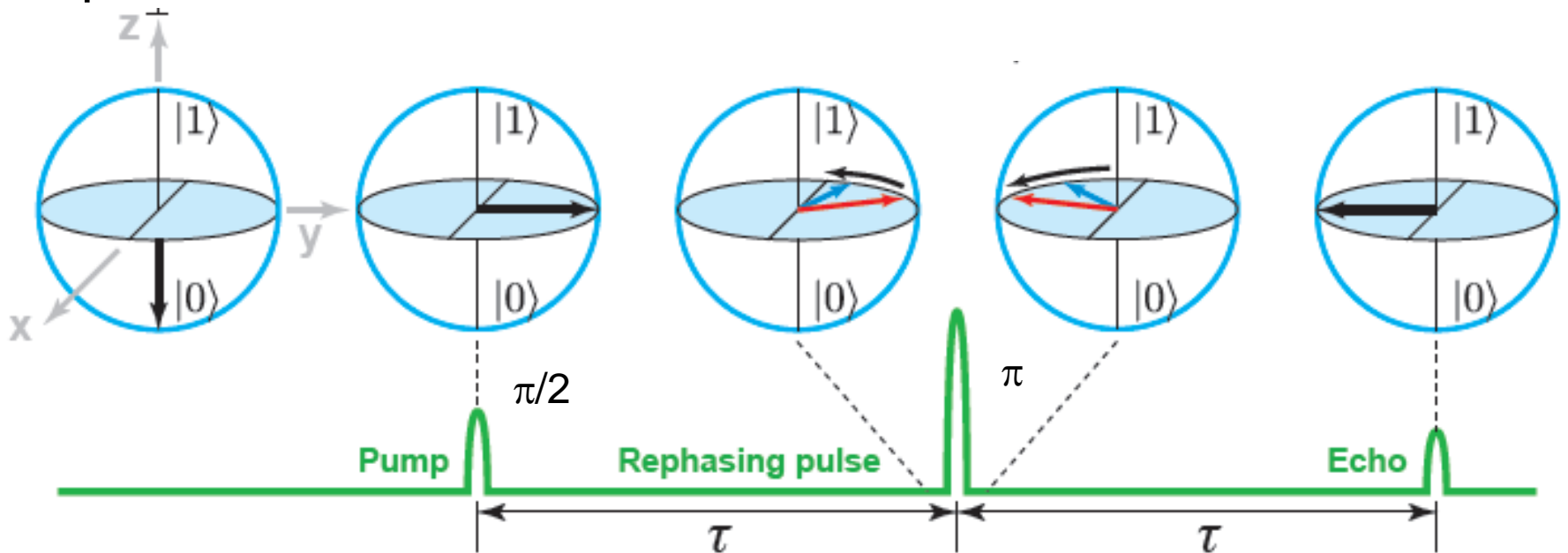


## The Bloch Sphere

H.M. Gibbs,  
 Phys. Rev. Lett. 29, 495 (1972),  
 Phys. Rev. A8, 446 (1973)

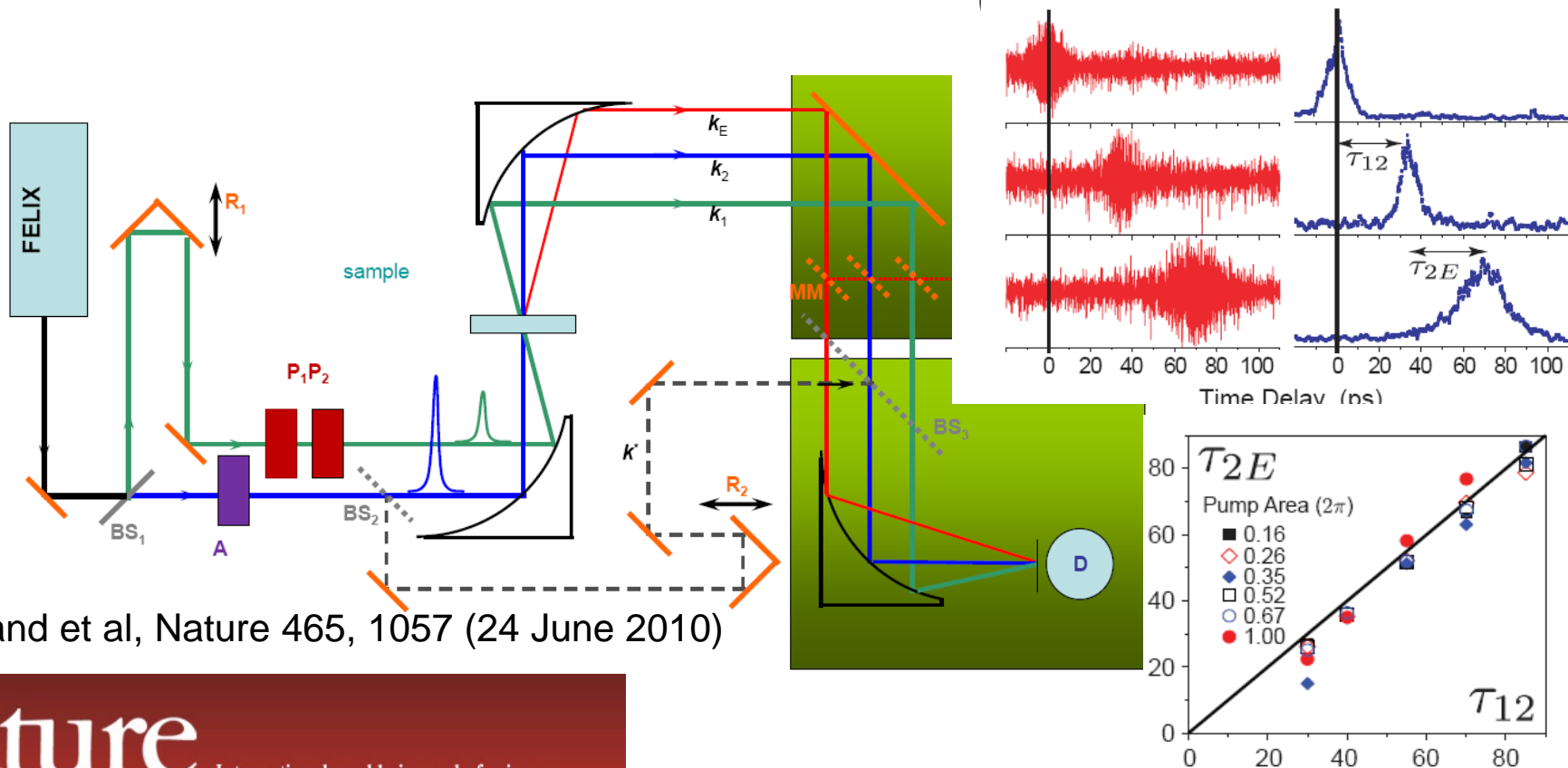
# The Hahn Echo

- The photon echo is analogous to the spin echo of ESR/NMR
- For an ensemble of oscillators with **inhomogeneity** in the natural frequency, a  $\pi$ -pulse can reverse the loss of phase



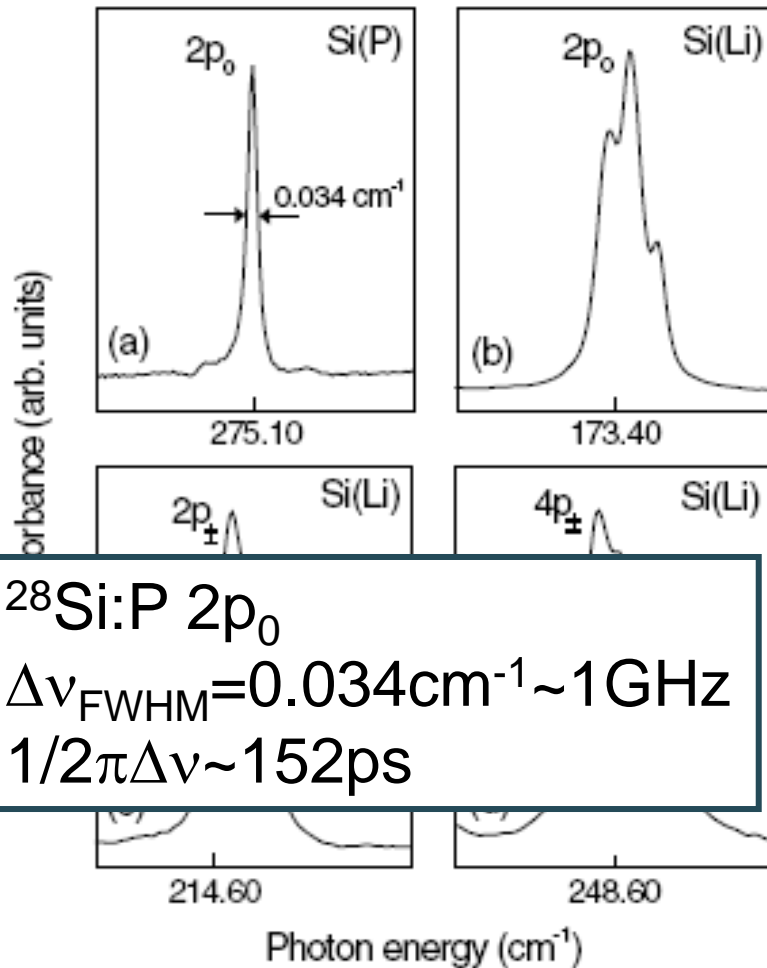
# Hahn echo in Si:P

- The arrival time of the echo is controlled by the delay between the pump and rephasing pulses
- Echo duration set by inverse inhomogeneous linewidth

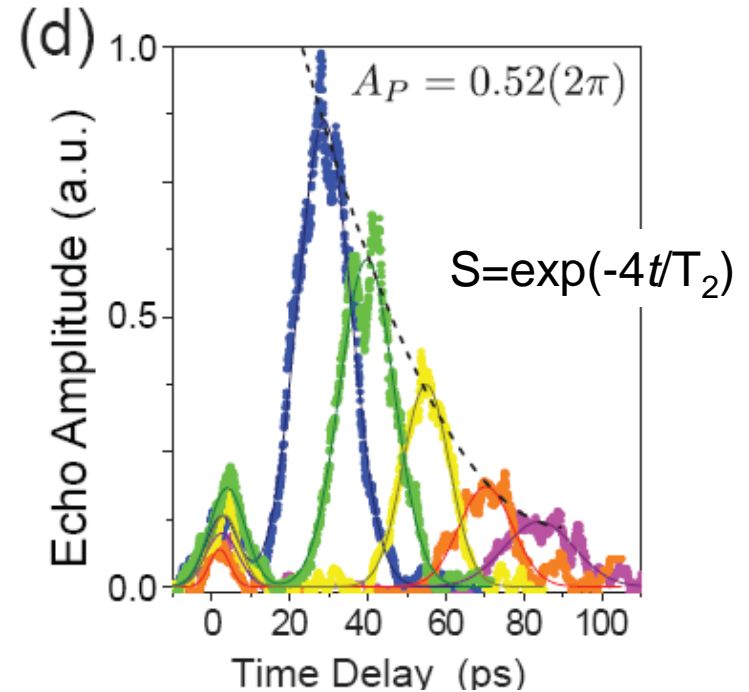




# Photon echoes for excited state $T_2$



$^{28}\text{Si:P } 2p_0$   
 $\Delta\nu_{\text{FWHM}} = 0.034 \text{ cm}^{-1} \sim 1 \text{ GHz}$   
 $1/2\pi\Delta\nu \sim 152 \text{ ps}$



Greenland et al,  
 Nature 465, 1057 (24 June 2010)

THz induced resonant charge transfer between donors

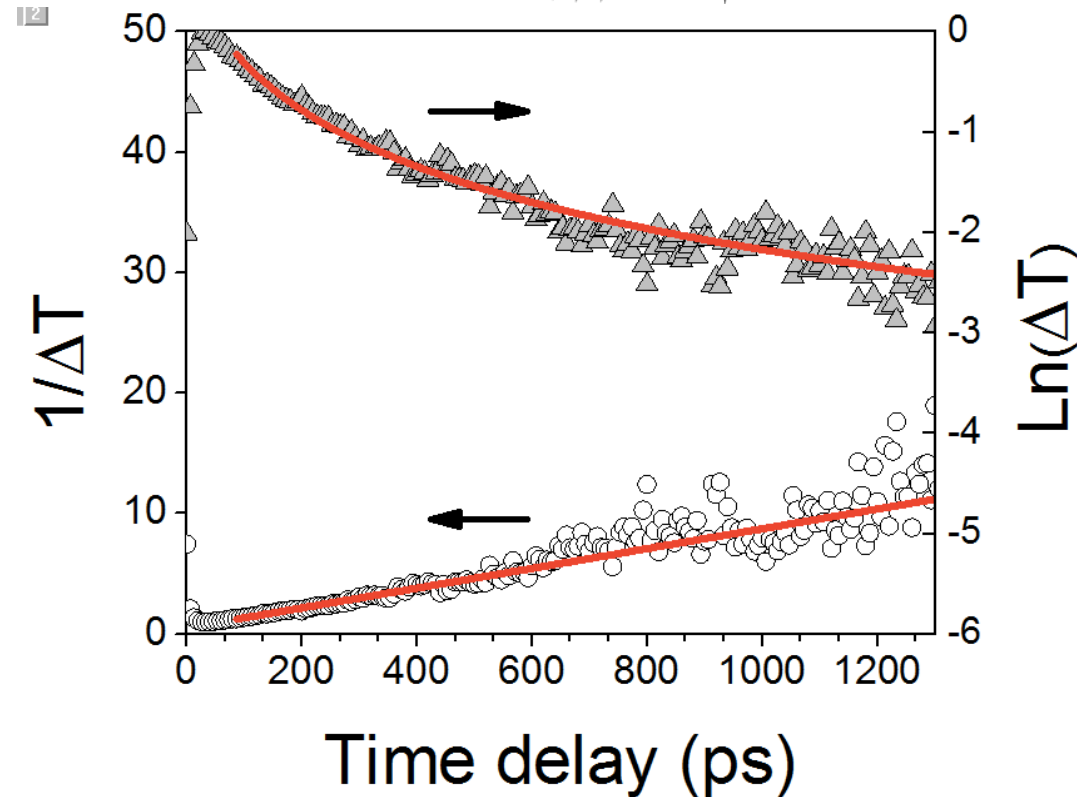
- Recombination from the continuum results in a reciprocal decay

$$\frac{dN_e}{dt} = -RN_e N_{D^+} = -RN_e^2$$

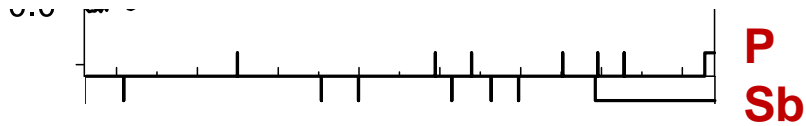
$$\frac{1}{N_e} - \frac{1}{N_e(0)} = Rt$$

- Whereas relaxation is exponential

$$\frac{dN_{2p}}{dt} = -\frac{1}{T_1} N_{2p} \quad \ln(N_{2p}) = -\frac{t}{T_1}$$



# Pumping to the continuum



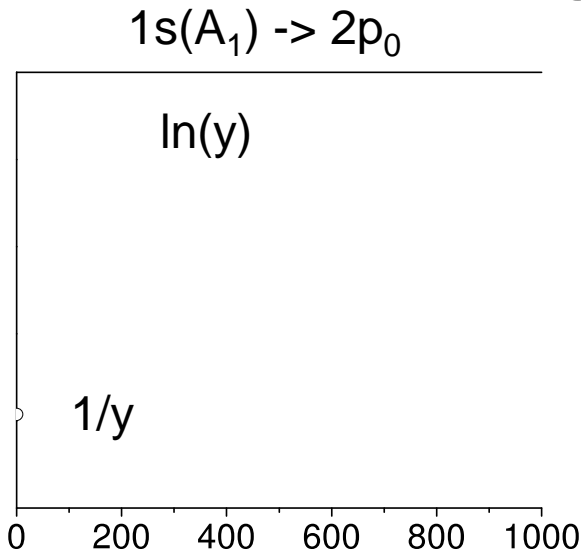
- ✓
- 1 -  $2p_0$ ,
  - 2 -  $2p_{\pm}$ ,
  - 3 -  $3p_0$ ,
  - 4 -  $3p_{\pm}$ ,
  - 5 -  $4p_{\pm}$ ,
  - 6 -  $5p_{\pm}$ ,
  - 7 - cb

- Samples all mounted at the same time on cold finger of LHe cryostat at base temperature, for pump-probe experiments

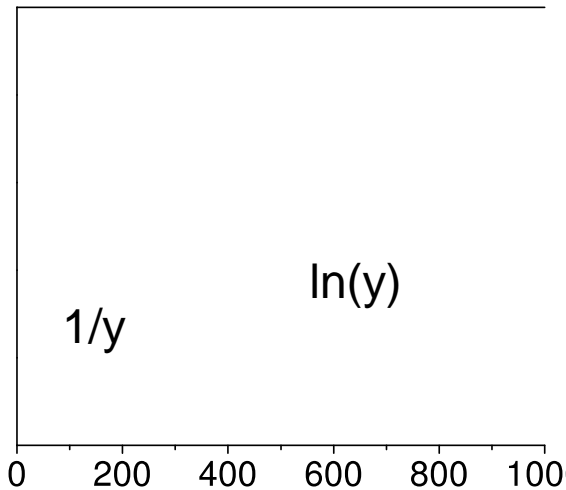
- Co-doped sample
  - P,  $1.8E15 \text{ cm}^{-3}$
  - + Sb,  $1.2E15 \text{ cm}^{-3}$
- Control samples, (monodoped)
  - P,  $4.0E15 \text{ cm}^{-3}$
  - Sb,  $3.8E15 \text{ cm}^{-3}$

## Co-doped sample

Si:P

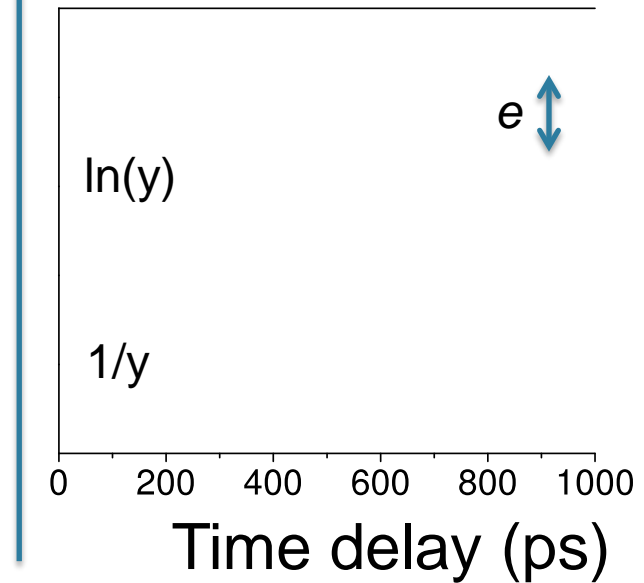


1s(A<sub>1</sub>) -> 2p<sub>±</sub>



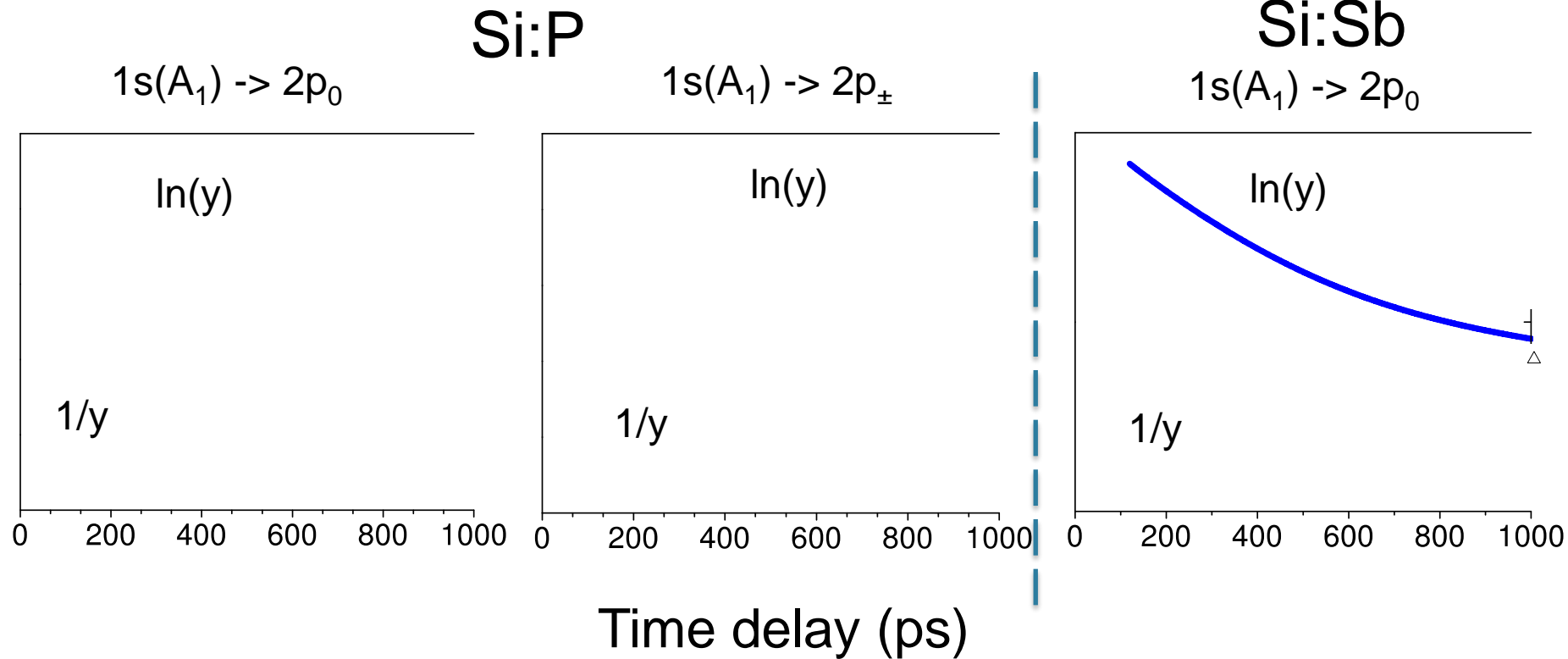
Si:Sb

1s(A<sub>1</sub>) -> 2p<sub>0</sub>



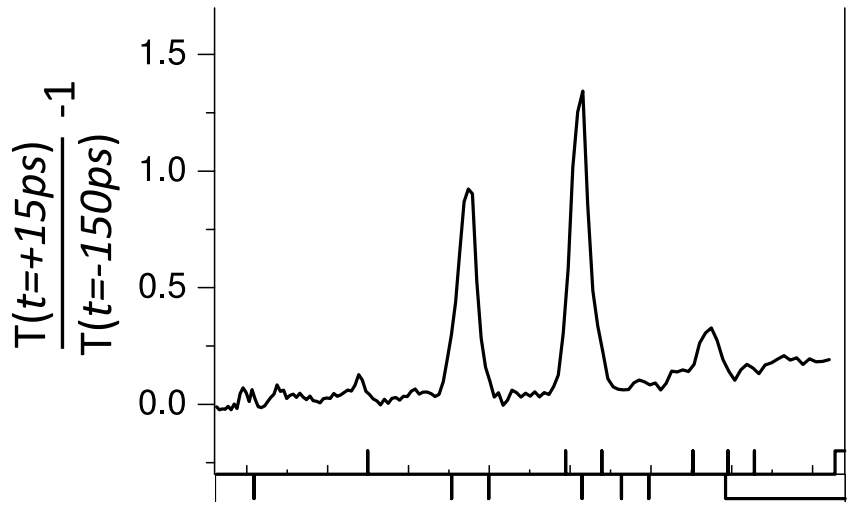
- Simple exponential decays
- The codoped sample was mounted at the same time and translated into the beam...

Resonant pumping in  
**monodoped control** samples



- Always get some combination of reciprocal and exponential
- Reciprocal contribution is ~10%

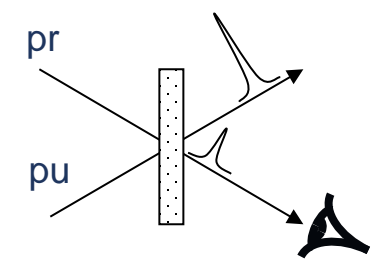
**Resonant pumping in codoped samples**



**P**  
**Sb**



- 1 -  $2p_0$ ,
- 2 -  $2p_{\pm}$ ,
- 3 -  $3p_0$ ,
- 4 -  $3p_{\pm}$ ,
- 5 -  $4p_{\pm}$ ,
- 6 -  $5p_{\pm}$ ,
- 7 - cb



- Co-doped sample
  - P,  $1.8 \times 10^{15} \text{ cm}^{-3}$
  - + Sb,  $1.2 \times 10^{15} \text{ cm}^{-3}$

Pump-probe signal is resonant

- Thomas et al PRB 23 5472 (1981)
- Concentration broadening
  - $D_{1s}D_{2p} \rightarrow D_{1s}D_{2p}$  transitions
- Appearance of "charge transfer states"
  - $D_{1s}D_{1s} \rightarrow D^+D^-$  transitions

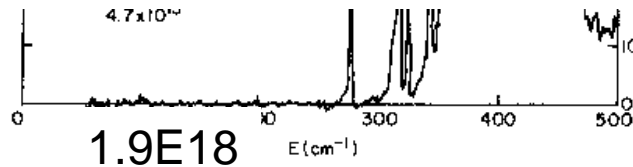


FIG. 2. Overview of the absorption coefficient (normalized to  $n_D$ ) as a function of photon energy for three separated donor densities  $n_D$  in samples of Si:P at 2 K. The three curves illustrate regimes of low, intermediate, and larger cluster absorption. At  $n_D = 4.7 \times 10^{15} \text{ cm}^{-3}$ , sharp lines of isolated donors, then at higher densities asymmetric broadening on the low-energy side of these lines due to donor pairs and finally at  $n_D = 4.7 \times 10^{17} \text{ cm}^{-3}$  almost the entire energy range due to clusters.

A. Experimental procedures

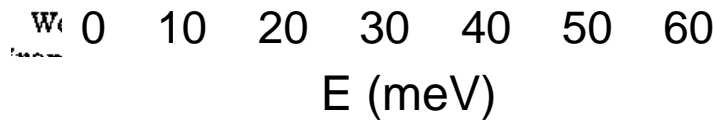


FIG. 3. Comparison of pair band absorption at two densities with the same variables plotted as in Fig. 7. The shaded areas are experimentally observed shifts which in turn define the values of the interatomic radii, e.g.,  $R_{D_{1s}D_{2p}}$  as indicated. The experimental results, solid circles, are compared with theoretical shape curves, dashed lines and their sum, solid line, on a semilogarithmic scale.

# Pair energetics

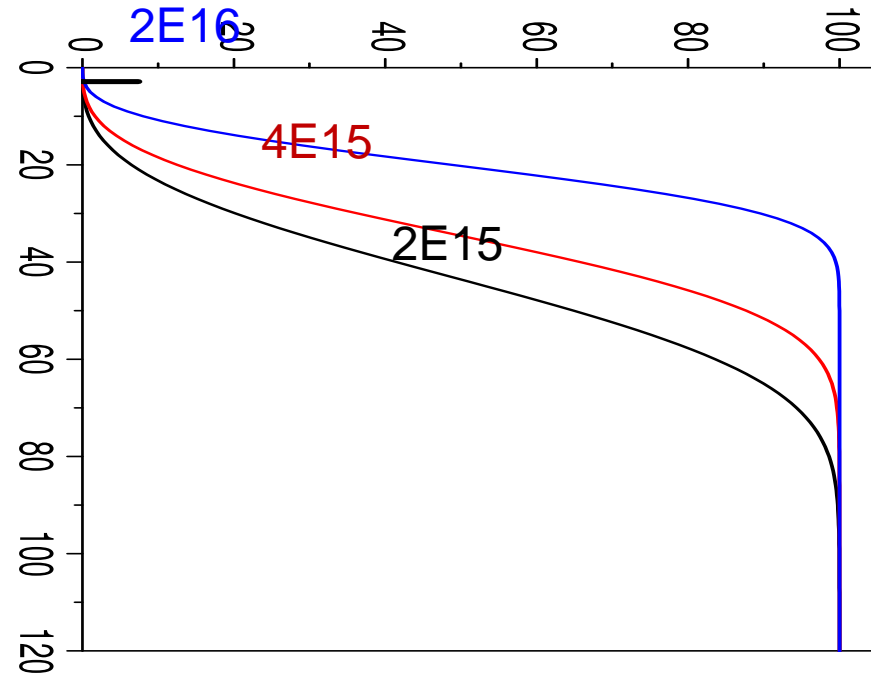


- P(no donors in vol  $V$ )  
=  $e^{-nV}$

- cumulative

probability

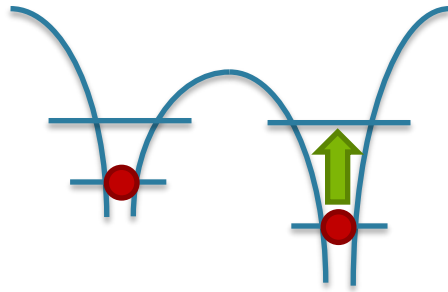
$$P(r) = 1 - \exp\left(\frac{-4\pi nr^3}{3}\right)$$



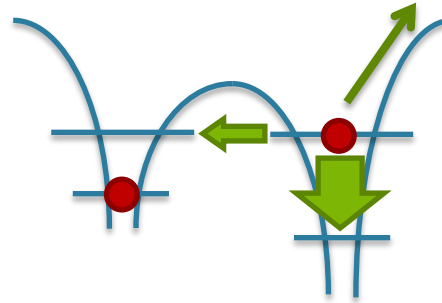
$$p(r)dr = 4\pi nr^2 \exp\left(\frac{-4\pi nr^3}{3}\right) dr$$

$$\langle r \rangle = 0.554n^{-1/3}$$

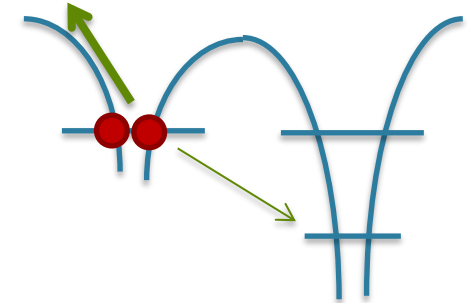
Poisson statistics of  
randomly doped samples



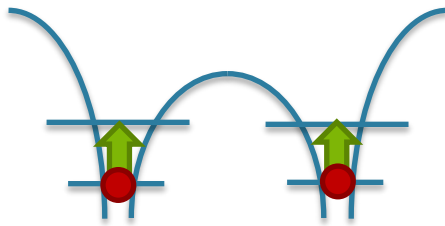
Resonant  
1s-2p  
pumping



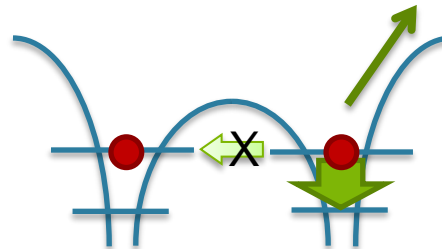
Resonant  
D+D-  
tunnelling,  
branching  
ratio 1:10



Relaxation is  
slower than  
thermal  
ionisation



Resonant  
1s-2p  
pumping

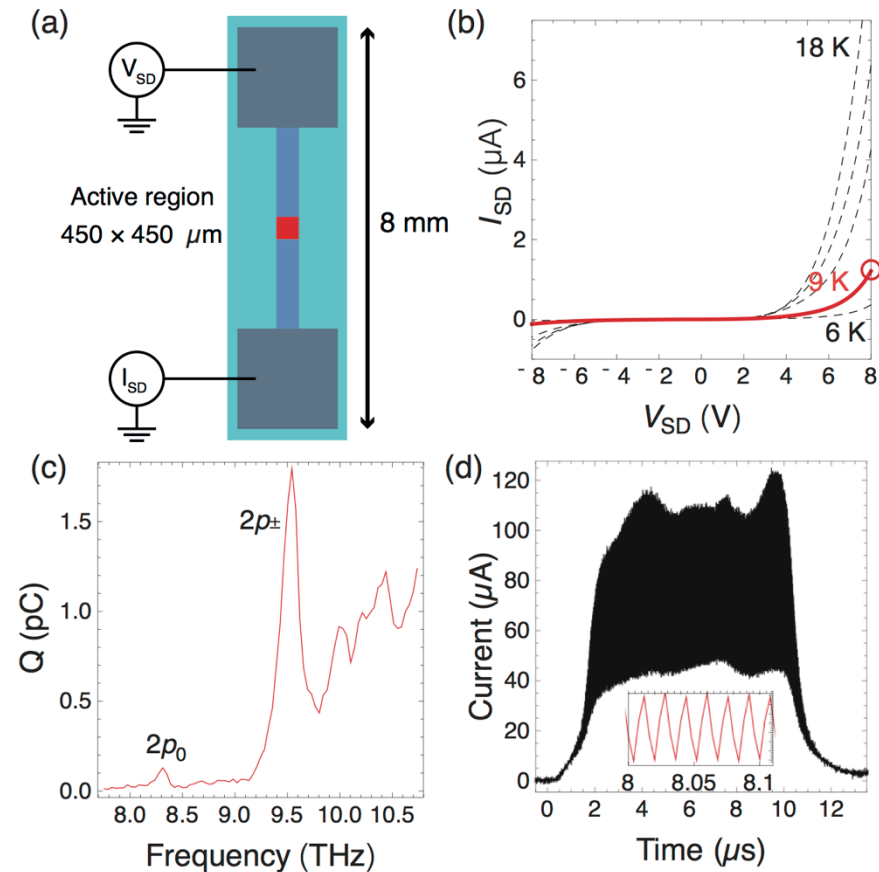


Resonant D+D-  
tunnelling is blocked if  
neighbour is excited

# conclusions

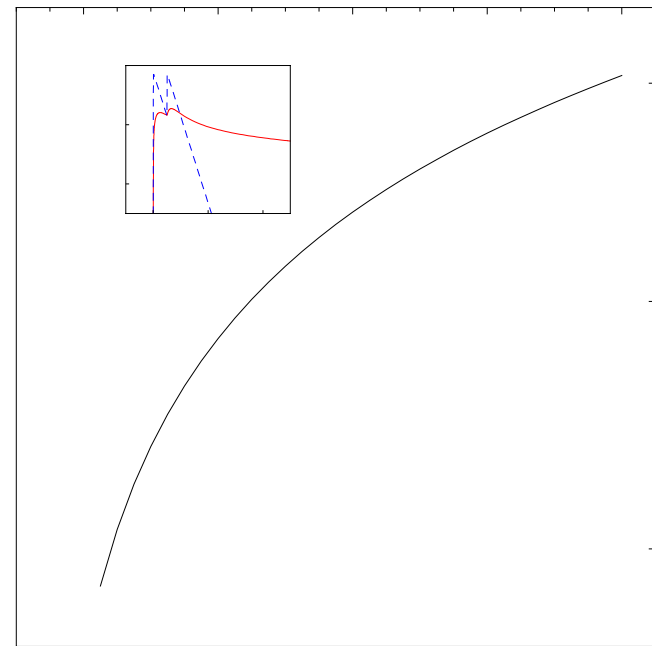
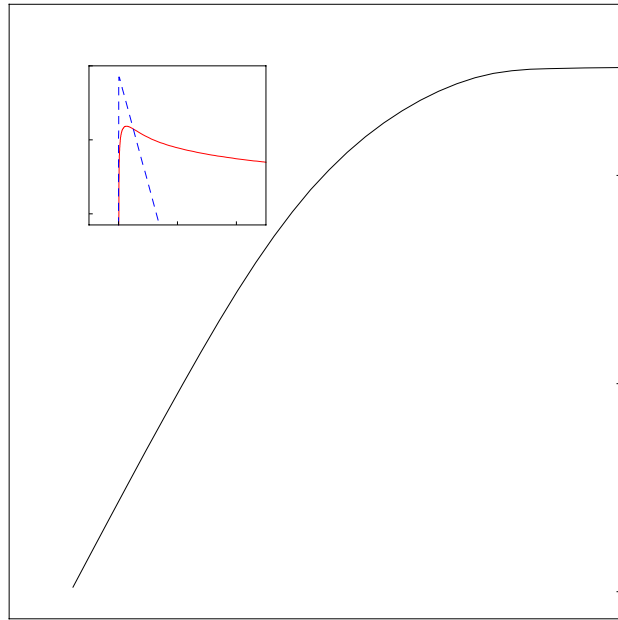
# Electrical readout

- Thermally excited electrons can be useful for readout



# Excitation dynamics

- Results can tell us the recombination rate coefficient  $R = 1.3 \pm 0.3) \times 10^{-11} \text{ m}^{-3} \text{ s}^{-1}$
- Now current can be used to calibrate the excitation



$\langle \Delta \rangle (x)$

# Si:P as a quantum information scheme

