Isotopically Engineered Silicon for Quantum Information Technology

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Major Collaborators:
Si growth: E. E. Haller (LBNL), H. Riemann (IKZ, Berlin)
Optical spectroscopy: M. L. W. Thewalt (Simon Fraser), A. Ramdas (Purdue)
Spin spectroscopy: A. M. Tyryshkin (Princeton), S. A. Lyon (Princeton), J. J. L. Morton (Oxford)

Precursor purchase: Initiatives for Proliferation Prevention program, Office of Nuclear Nonproliferation (NN-40), US Department of Energy
Scientific development: Basic Energy Sciences, Office of Science, US Department of Energy

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Why silicon quantum computing?

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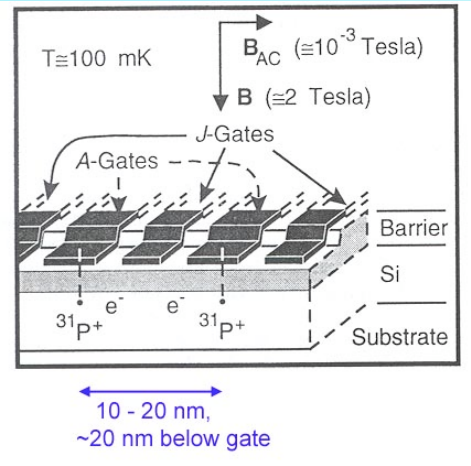

B. E. Kane, Nature **393**, 133 (1998)
>1600 cites per Google Scholar…

- >50 years of Si technology
  - Large scale growth of dislocation-free crystals
  - Dopants and metals controlled to <<ppt
- But how "perfect" is Si, really?
  - 7.8% mass defects!
  - ^{28}Si: 92.2%
  - ^{29}Si: 4.7%
  - ^{30}Si: 3.1%

Isotopes and spins in Si

Not a new idea…

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G. Feher, J. P. Gordon, E. Buehler, E. A. Gere, and C. D. Thurmond, *Phys. Rev.* **109**, 221 (1958)

In the present experiments, the spin resonance, which is inhomogeneously broadened by hyperfine interactions of the donor electrons with the Si^{29} nuclei,[2] was narrowed from 2.7 oersteds in width to 0.22 oersted through the use of a crystal of isotopically purified silicon[3] [estimated final isotopic purity $(99.88 \pm 0.08)\%$ Si^{28}]. As a result the oscillation condition was easily satisfied.

J. P. Gordon and K. D. Bowers, *Phys. Rev. Lett.* **1**, 368 (1958)

With the introduction of the spin-echo technique by Hahn,[1] it became possible to observe and measure directly transverse relaxation times T_2 in substances where the resonance lines involved were inhomogeneously broadened by local field variations. The spin-echo technique has

Table I. Experimental results.

Sample	Type	Concentration of donors (cm^{-3})	Spin-lattice time T_1 (sec)	Phase memory time T_2 (sec)
1.	P donors	3×10^{16}	~100	2.4×10^{-4}
2.	P donors	10^{17}	~1	2.0×10^{-4}
3.	Li donors	3×10^{16}	~100	2.4×10^{-4}
4.	P donors in isotopically enriched Si^{28}	4×10^{16}	~50	5.2×10^{-4}

500 μs

Challenges in making isotopically controlled crystals and structures

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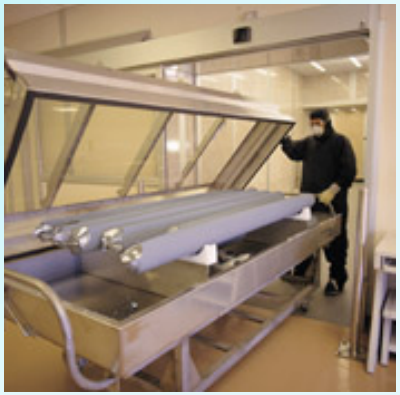
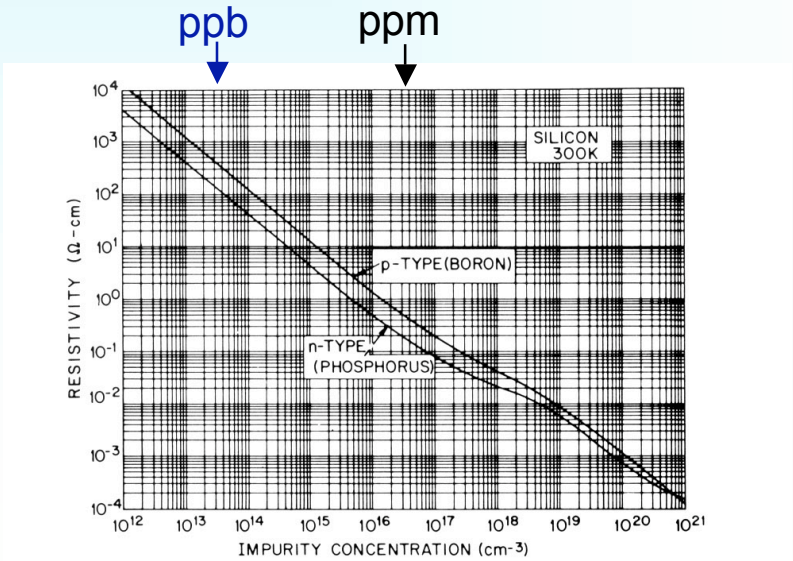
- Si chemical purity (metals, dopants, etc.) improves with increasing production scale



- In chemical terms, impurity requirements are stringent



So, how is isotopically enriched Si made?

To retain respect for sausages and laws, one must not watch them in the making

Attrib. to O. von Bismarck

The enrichment step

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- Enriched SiF_4 from Russia
  - >99.9% ^{28}Si
  - >90% ^{29}Si
  - >90% ^{30}Si

  formerly provided 40% of Soviet Union capacity for uranium enrichment

Major impurities: CF_4, C_2F_6, CO_2, Ar, O_2, HF, H_2O at up to 50 ppm

Electrochemical Plant (ECP)
Krasnoyarsk-45 / Zelenogorsk

Silane production

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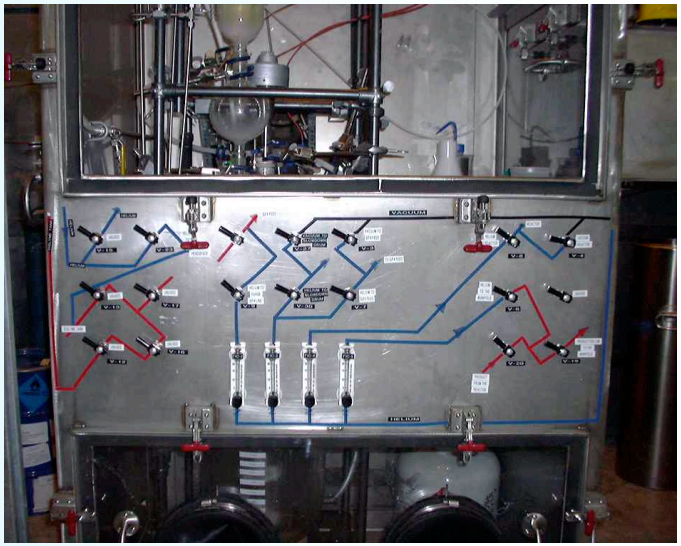
- Voltaix Inc (NJ, USA)
  - In 150 g batches react SiF_4 with $LiAlH_4$ in diglyme solution to produce SiH_4 (silane)
  - Condense silane on molecular sieve

  Major detectable impurities:
  - CF_4 and CH_4 up to 100 ppm

  Test epi wafers:
  - From 5-10 Ω-cm to >1000 Ω-cm (highest >3000 Ω-cm)



LBNL production of poly-Si

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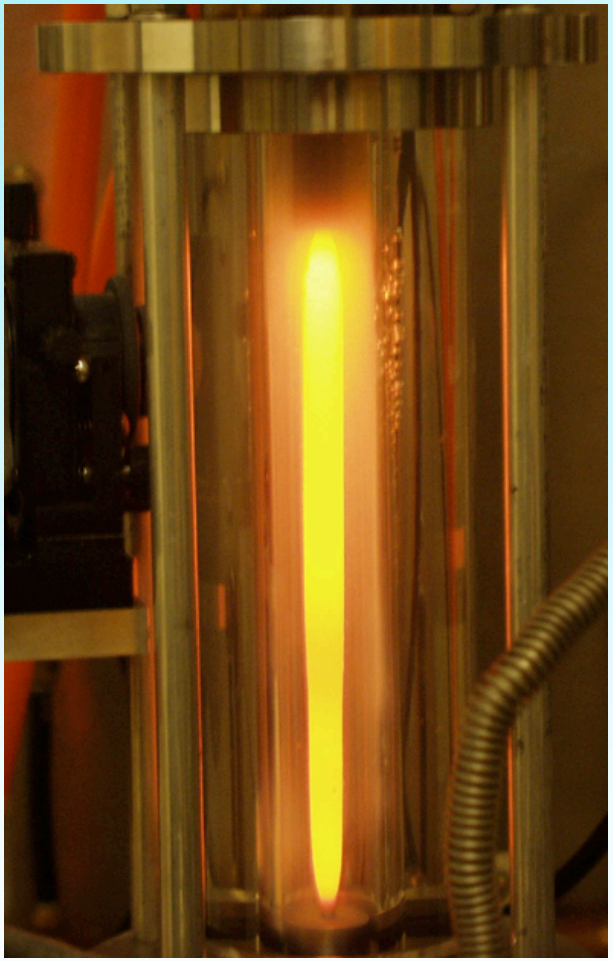
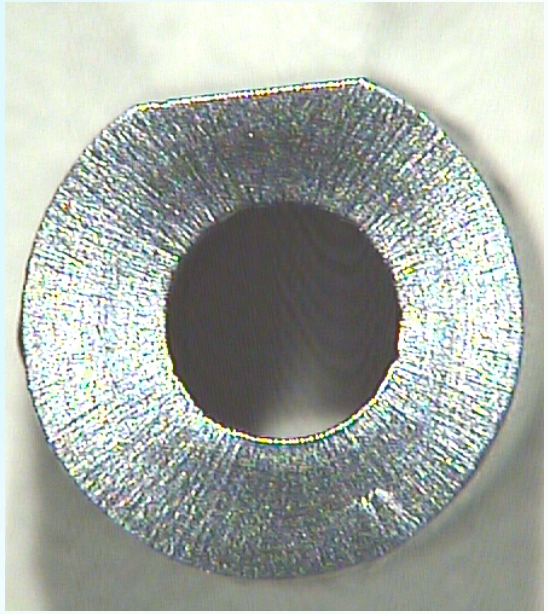


Poly-Si rod growing at 730°C
in a recirculating flow of 2% $^{28}SiH_4$
in hydrogen

High yield and high chemical purity



12 mm diameter poly-Si in cross section
with 3 mm graphite starter rod removed

95% yield

Single crystal silicon enriched in all three stable isotopes

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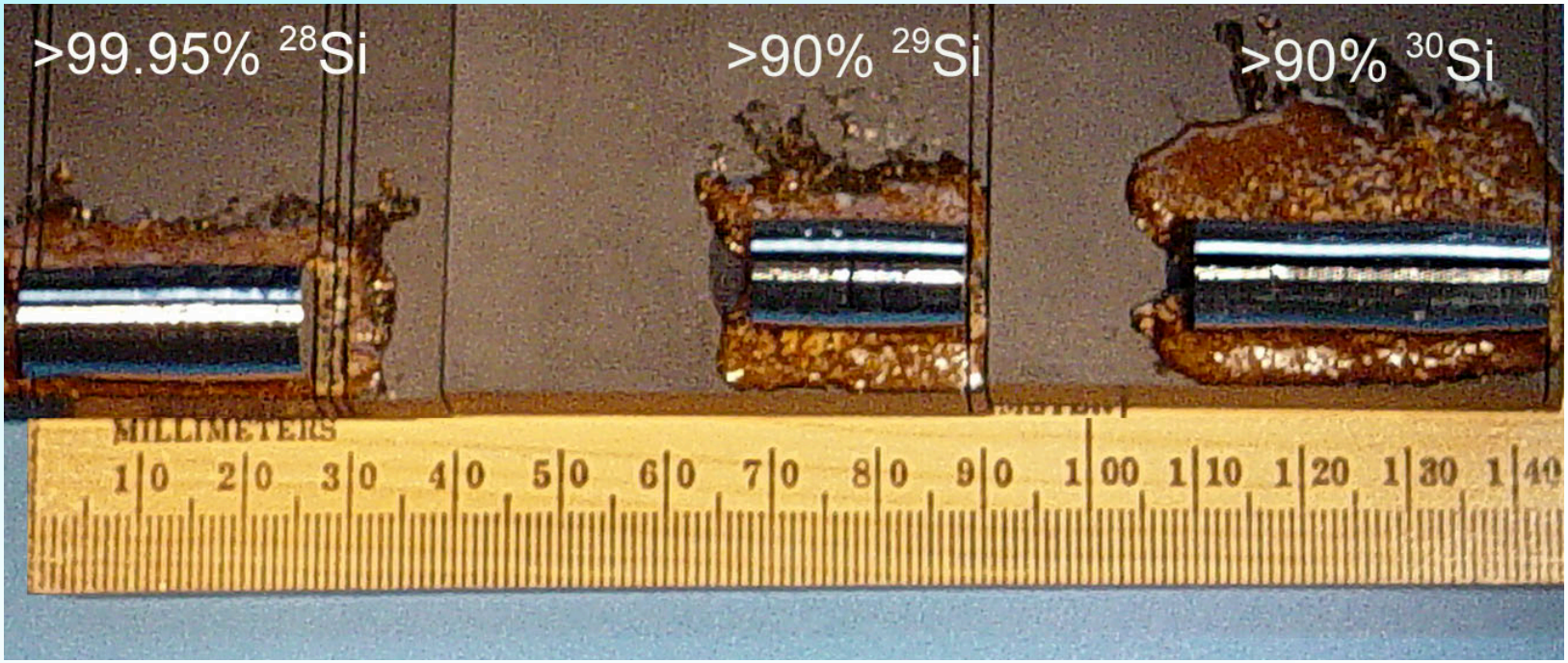
Floating zone crystal growth by Institute for Crystal Growth (Berlin)



Electrically active impurities:

P and B as low as mid-10^{13} cm^{-3}

Other impurities:

C < 10^{16} cm^{-3}, O < 10^{15} cm^{-3}

J. W. Ager III, J. W. Beeman, W. L. Hansen, E. E. Haller , I. D. Sharp, C. Liao, A. Yang, M. L. W. Thewalt, and H. Riemann, J. Electrochem. Soc **152**, G448-G451 (2005).

Not just for quantum computing

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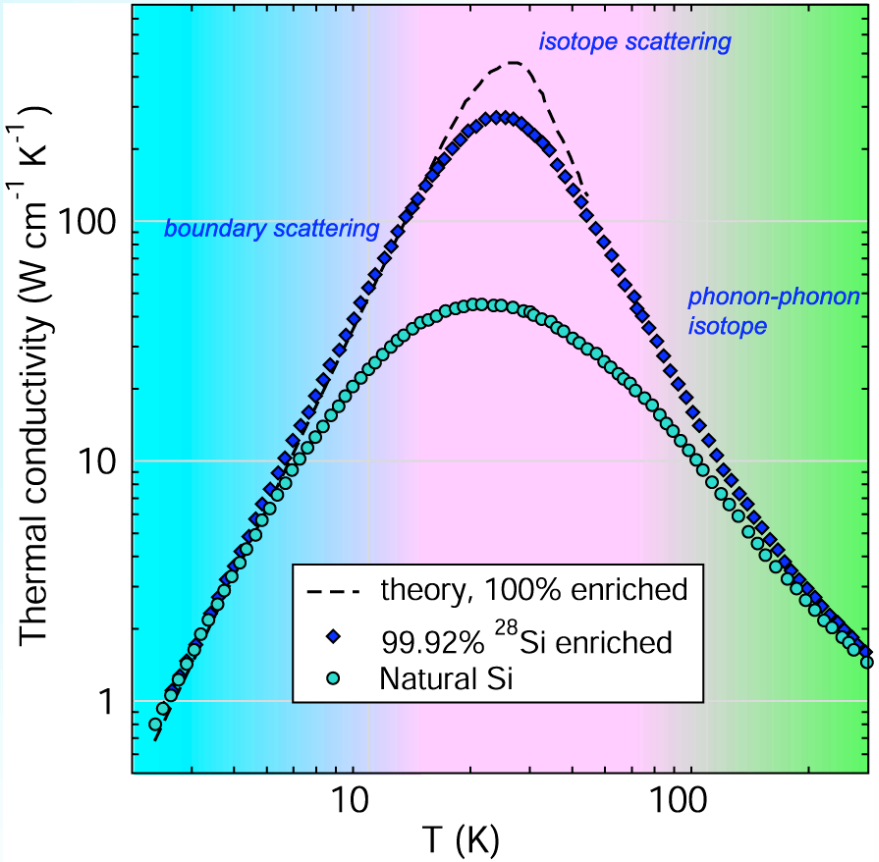
- Thermal transport
- Near-surface diffusion

  PRL 97, 055503 (2006)
- Direct and indirect gaps vs. mass and the "true" Si band gap

  PRB 70, 193201 (2004)

  PRB 72, 153203 (2005)
- Optical detection of hyperfine splitting of exciton bound to ^{31}P

  PRL 97, 22740 (2006)



Isotope effects

Band structure

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Variation of band gap E_g with average isotopic mass M

$$\left(\frac{\partial E_g}{\partial M}\right)_{T,P} = \left(\frac{\partial E_g}{\partial M}\right)_{T,V} + \frac{D}{V}\left(\frac{\partial V}{\partial M}\right)_{T,P}$$

Electron-phonon interaction

dominant effect in Si

Dependence of band gap on volume

D is the deformation potential

Both terms vary as $M^{-1/2}$

$$E_G(M) - E_G^{\infty} = -CM^{-1/2}$$

E_G^{∞} is the band gap for a static lattice

Band gap renormalization

High-purity material required

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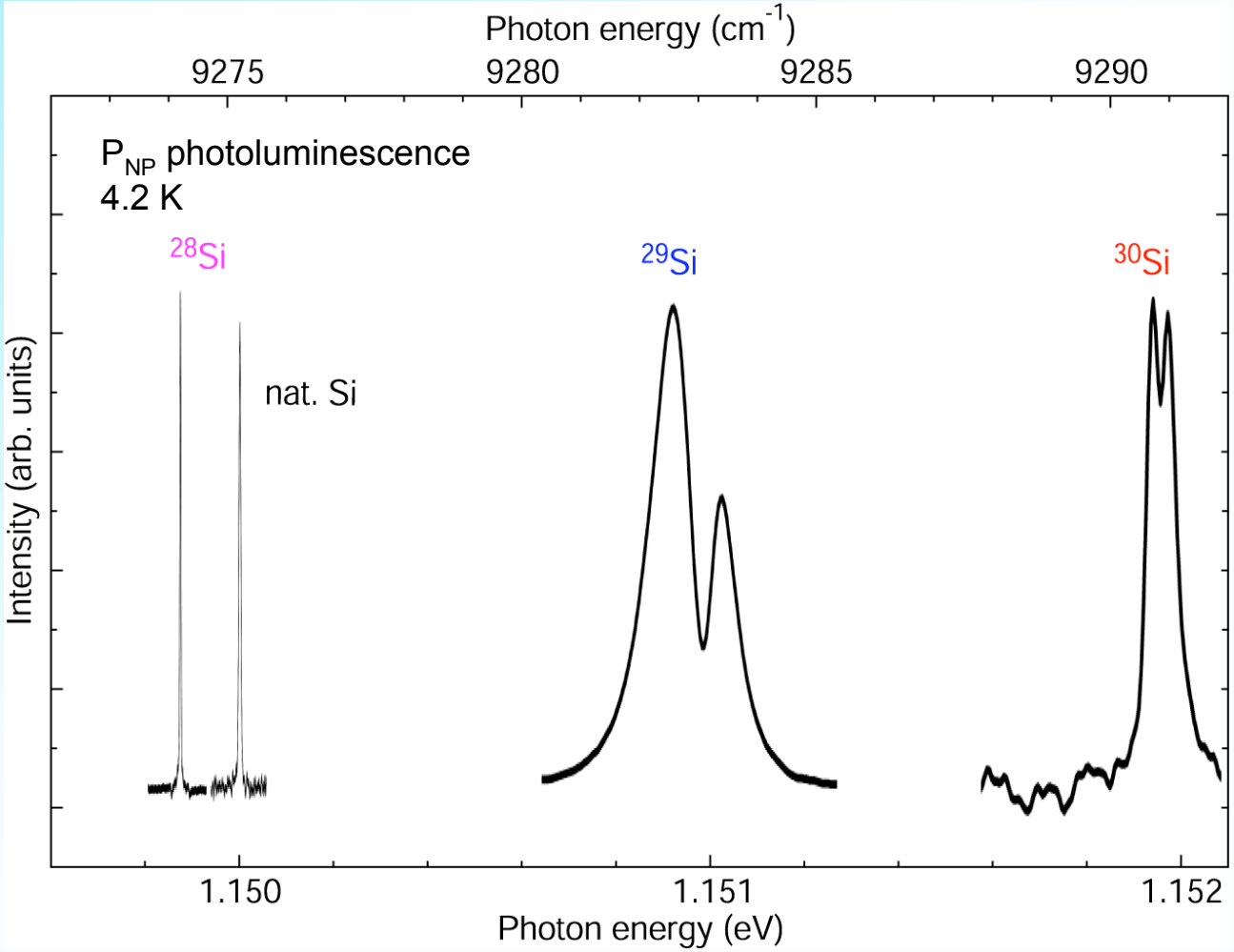

ca. 1 meV/amu renormalization observable but lines broadened, split, and shifted due to carbon contamination (>10^{16} cm^{-3})

Adapted from Karaiskaj *et al.*, Solid State Commun. 123, 87 (2002).

P_{NP}: narrow line for all isotopes

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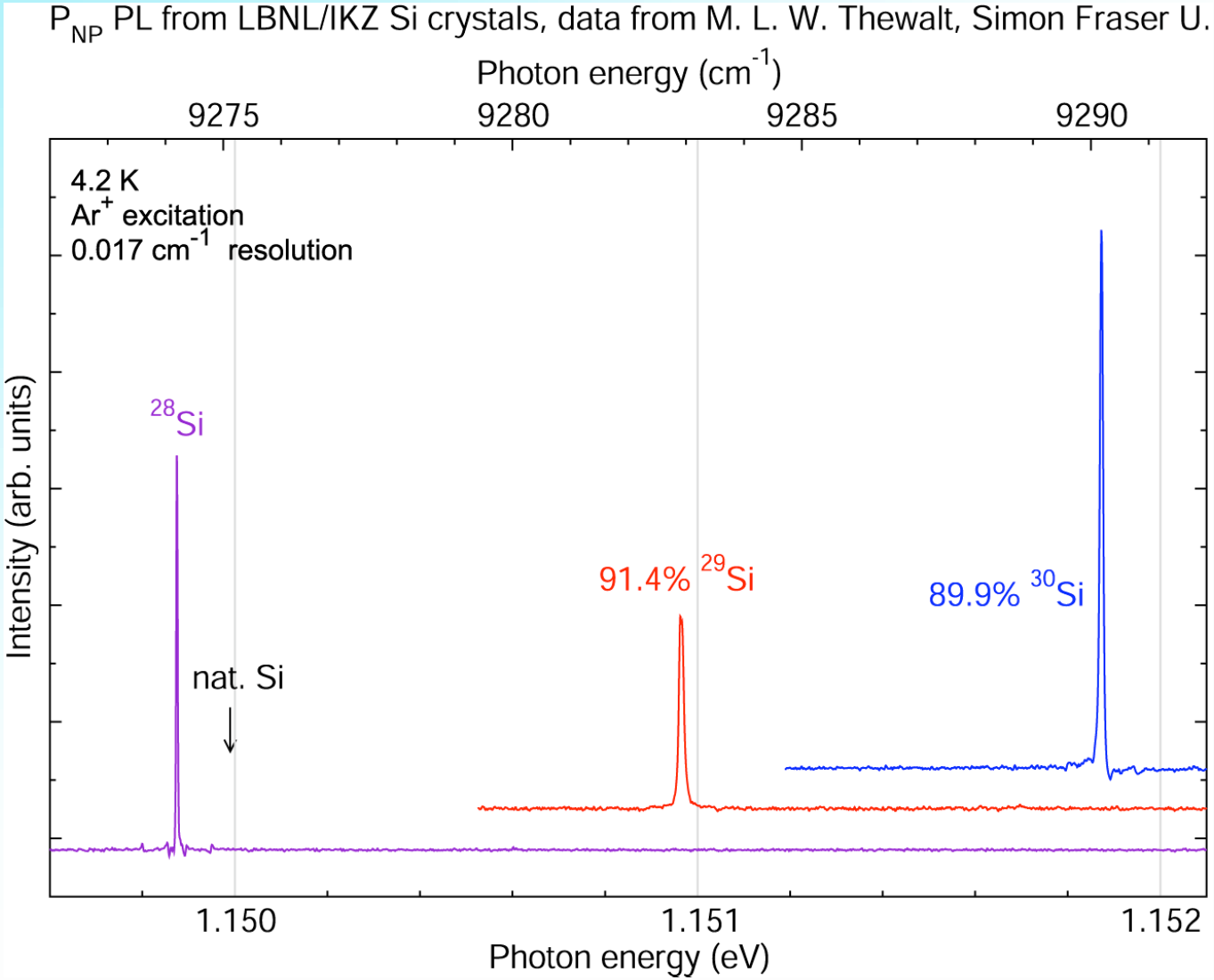

J. W. Ager III, J. W. Beeman, W. L. Hansen, E. E. Haller , I. D. Sharp, C. Liao, A. Yang, M. L. W. Thewalt , and H. Riemann, J. Electrochem. Soc **152**, G448-G451 (2005).

Si excitonic band gap
Precise measurement

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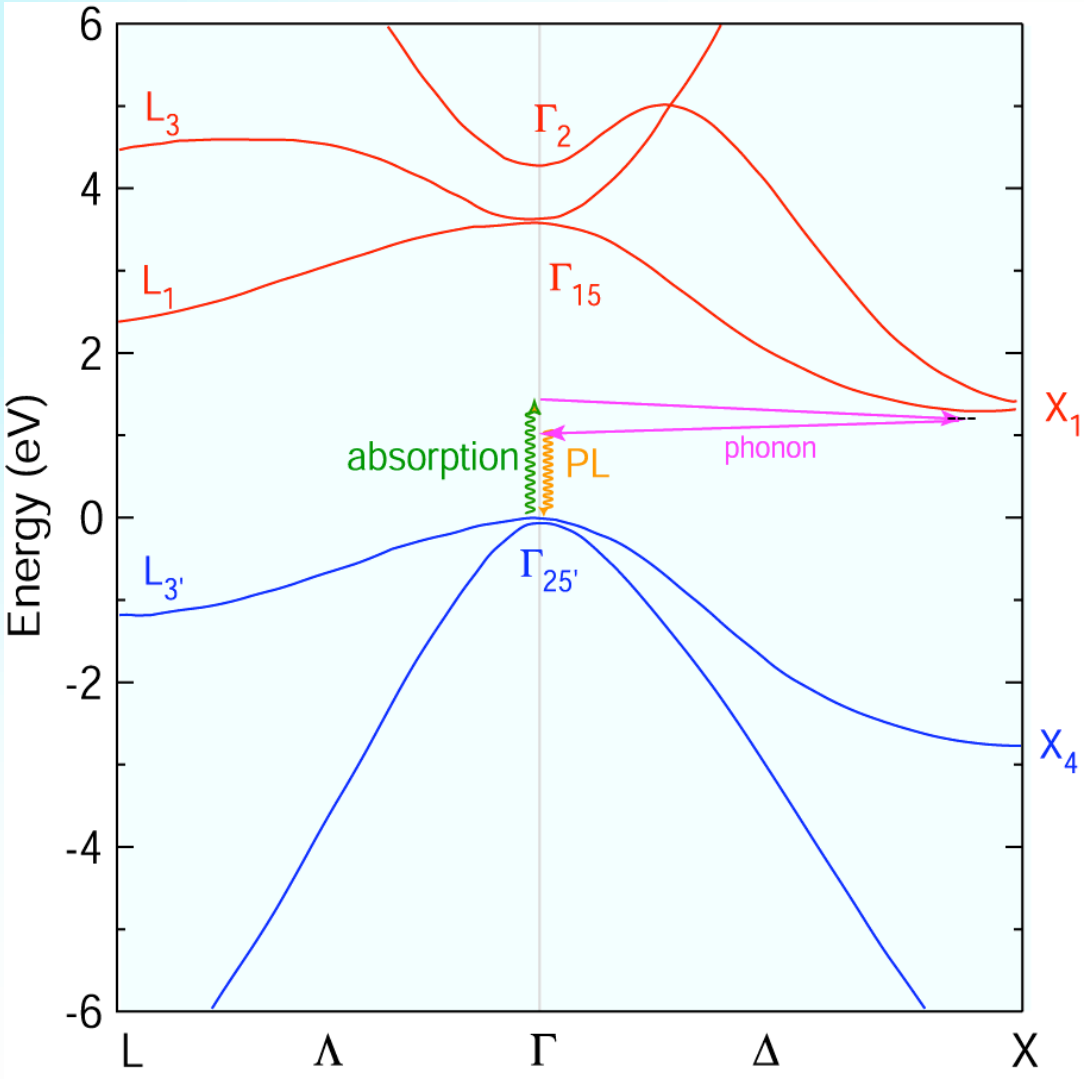
- Phonon-assisted transitions across indirect gap

  Absorption: E_{gx} + phonon

  PL: E_{gx} - phonon



Empirical pseudopotential band structure adapted from Cohen and Chelikowsky, *Electronic and Optical Properties of Semiconductors*, 1989

Fundamental Si bandgap

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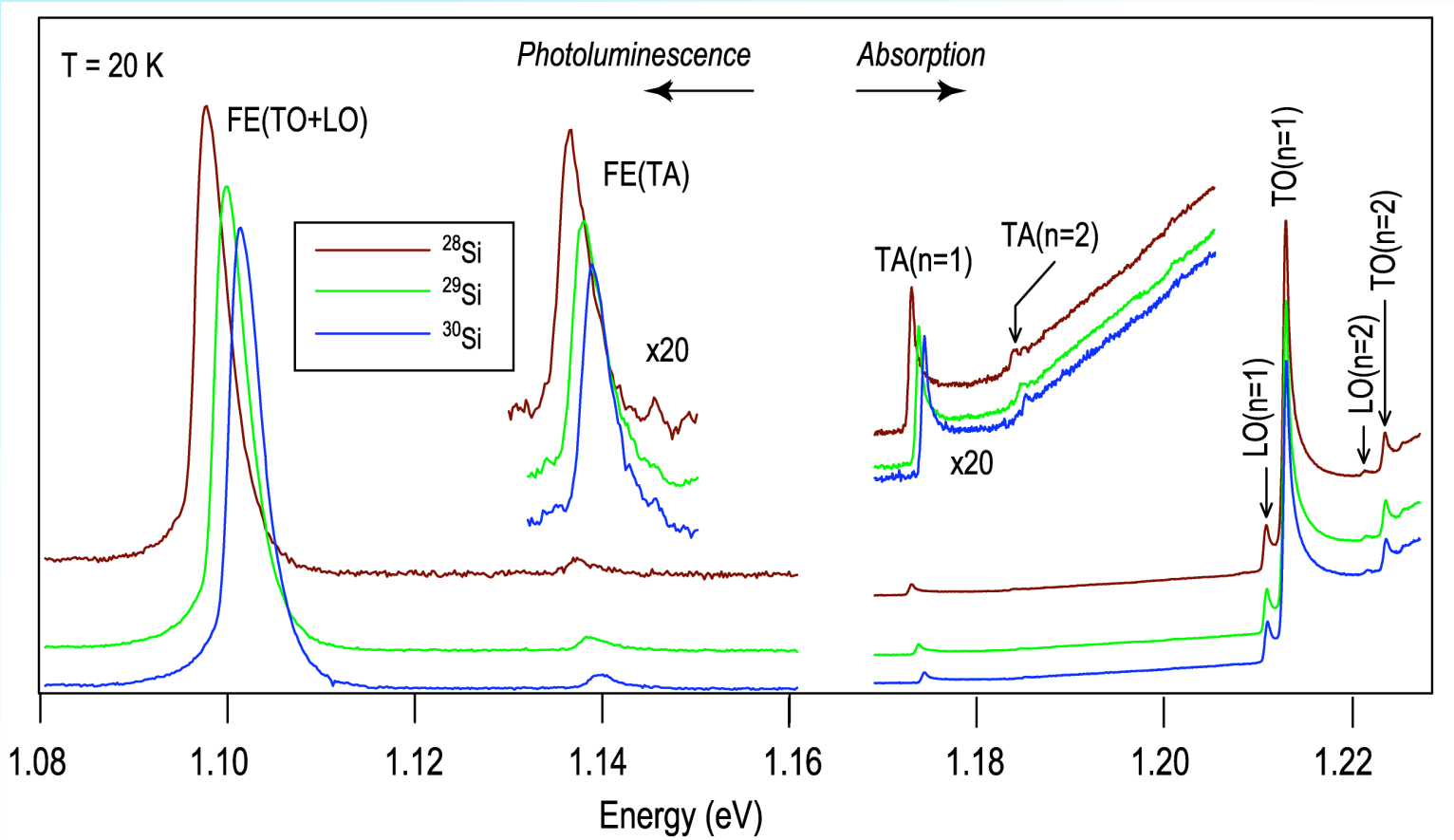
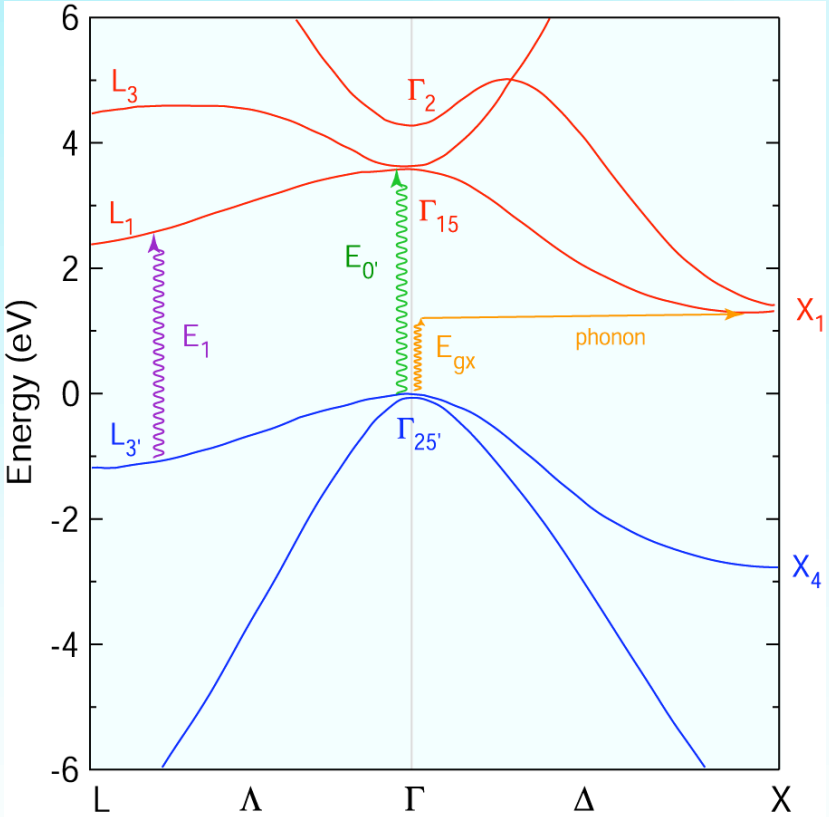
Optical absorption *and photoluminescence (PL) across the indirect gap of silicon occur with assistance from phonons (TA, LO, and TO where T = transverse, L = longitudinal, O = optical, and A = acoustic). Absorption peaks are observed at the band gap plus the phonon energy; PL peaks are at the band gap minus the phonon energy.*

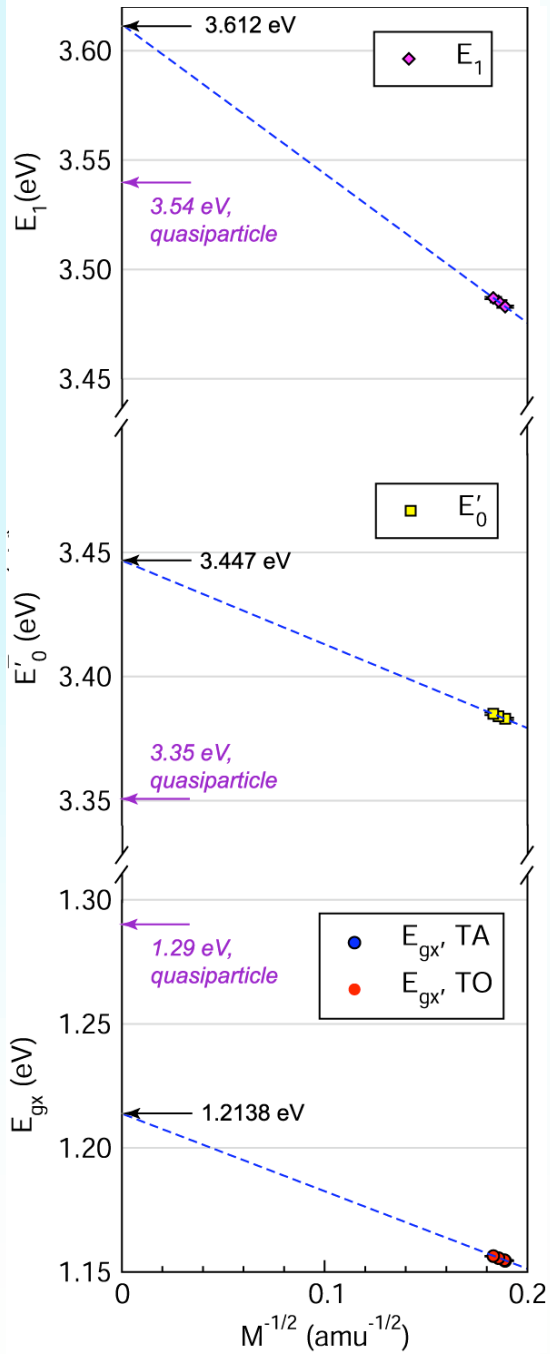


S. Tsoi, H. Alawadhi, X. Lu, J. W. Ager III, C. Y. Liao, H. Riemann, E. E. Haller, S. Rodriguez, and A. K. Ramdas, "Electron-phonon renormalization of electronic band gaps of semiconductors: isotopically enriched silicon," Phys. Rev. B **70**, 193201 (2004).

Purely electronic Si bandgaps

Direct and indirect





Quasiparticle band gaps from Hybertsen and Louie, PRB 34, 5390 (1986)

Tsoi, Alawadhi, Lu, Ager, Liao, Riemann, Haller, Rodriguez, and Ramdas, PRB **70**, 193201 (2004)
Tsoi, Rodriguez, Ramdas, Ager, Riemann, Liao, and Haller, Phys. Rev. B **72**, 153203 (2005).

Back to Si quantum computing

n-type, low 10^{15} cm^{-3}

Towards long T_2 …

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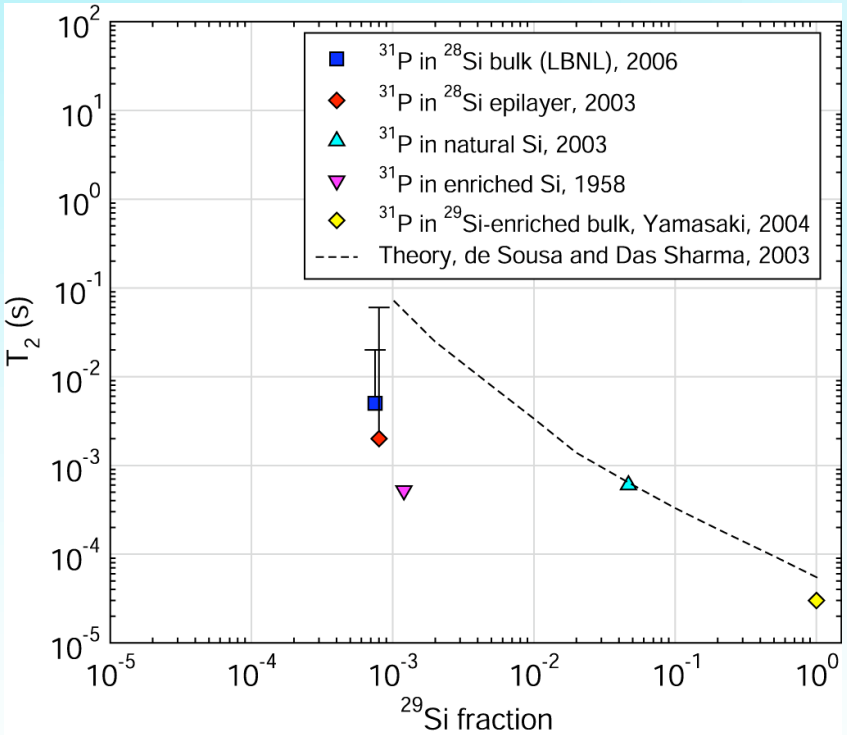
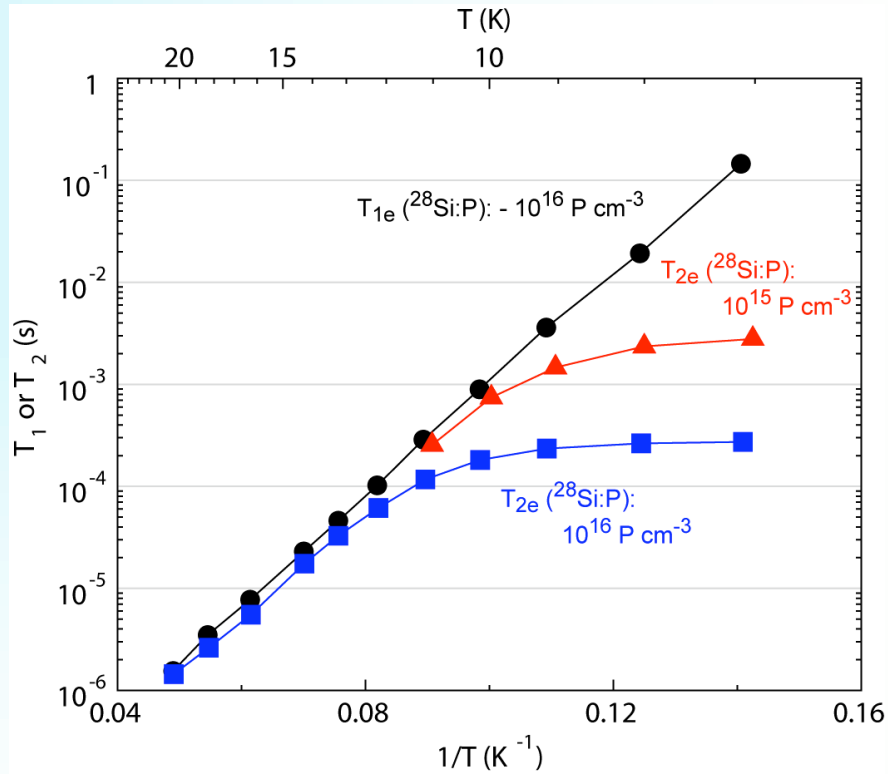
Of course removing ^{29}Si increases T_2



Reducing P content also helps



A. M. Tyryshkin, J. J. L. Morton, S. C. Benjamin, A. Ardavan, G. A. D. Briggs, J. W. Ager III, and S. A. Lyon, J. Phys. Cond. Mat. 18, S783 (2006).

Can the crystals be made better?

Controlling P concentration

Zone refining

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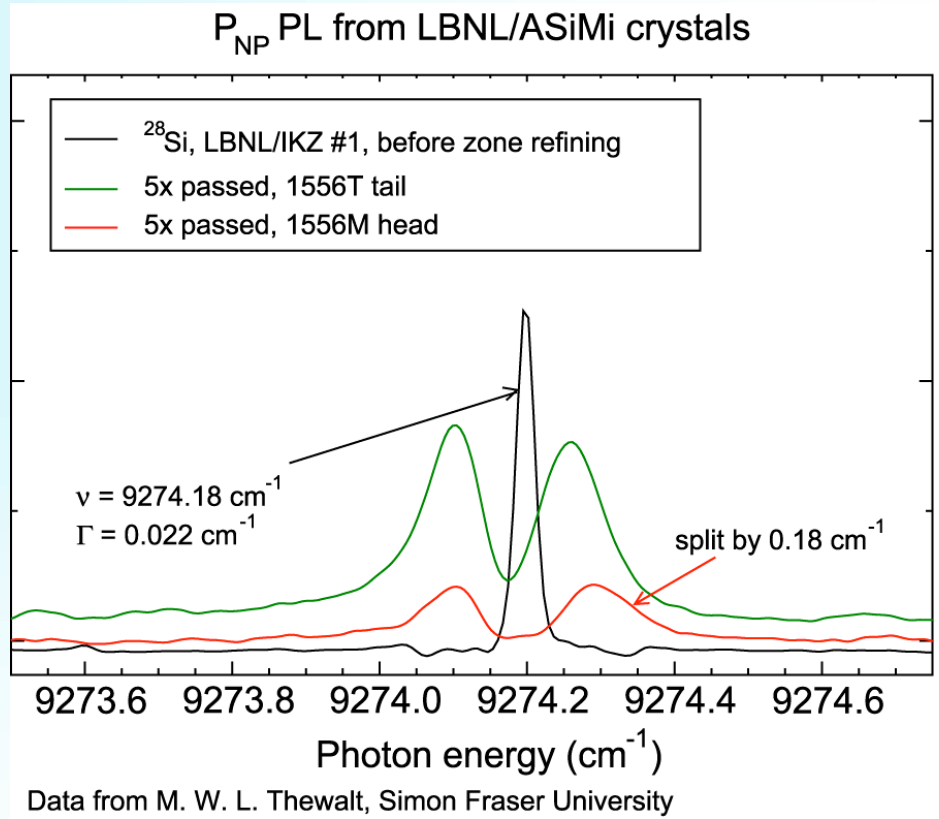
Five passes of zone refining (Advanced Silicon Materials) reduced impurity concentration

	P (cm^{-3})	B (cm^{-3})
Head	$8x10^{13}$	$6x10^{12}$
Tail	$4x10^{14}$	$1x10^{13}$

But carbon introduced at >10^{16} cm^{-3}



Controlling P concentration

Float zone regrowth by IKZ

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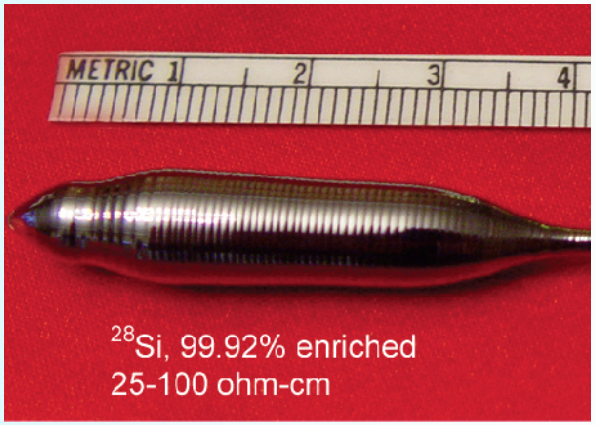
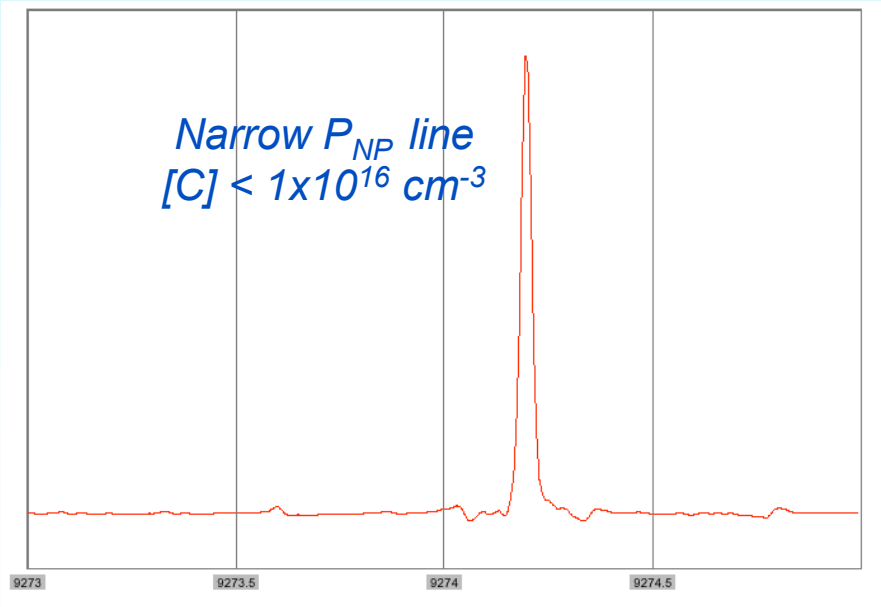
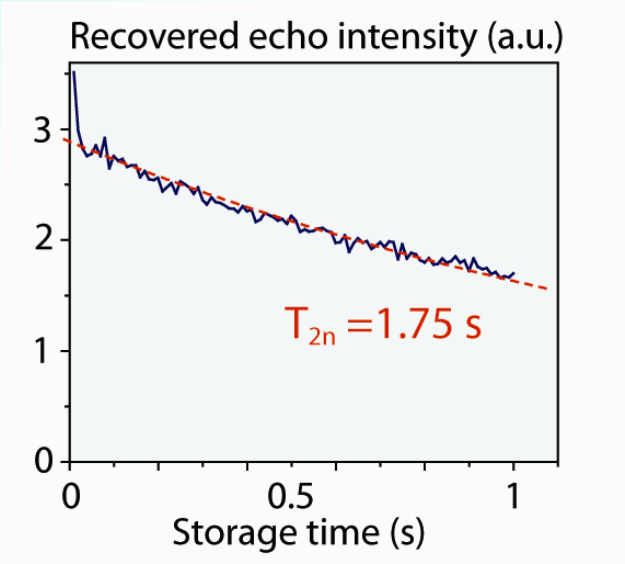
	P (cm^{-3})	B (cm^{-3})
Head	1.9×10^{14}	1.5×10^{13}
	1.8×10^{14}	7.2×10^{13}
	1.8×10^{14}	6.0×10^{13}
Tail	4.8×10^{14}	5.4×10^{13}







Morton *et al.*, Nature **455** 1085 (2008).

Conclusions and outlook

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- Dislocation-free Si bulk single crystals (and epilayers) with high isotope enrichment and low concentration of electrically active and other impurities can be made
  - Precursor purity is crucial

- This material underlies current and future breakthroughs in single spin control and high resolution spectroscopy in the solid state

Storing coherence on the nuclear spin

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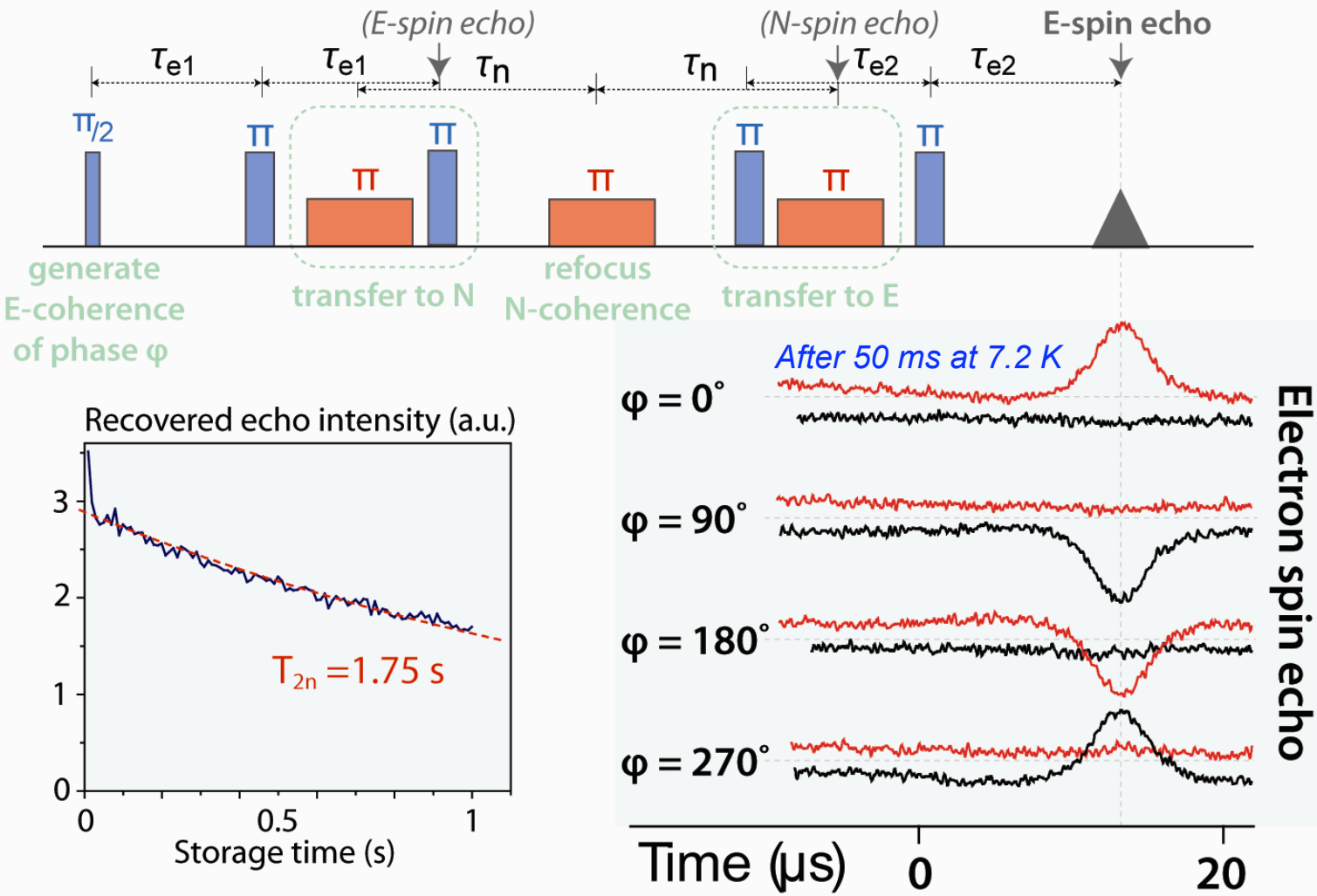

J. J. L. Morton, A. M. Tyryshkin, R. M. Brown, S. Shankar, B. W. Lovett, A. Ardavan, T. Schenkel, E. E. Haller, J. W. Ager, and S. A. Lyon, Nature **455** 1085 (2008).