Optically Detected Hyperfine Splitting & Optical Nuclear Spin Hyperpolarization of the ^{209}Bi Donor in ^{nat}Si

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*- Morley group - Phys. Rev. Lett. **105**, 067602 (2010)*

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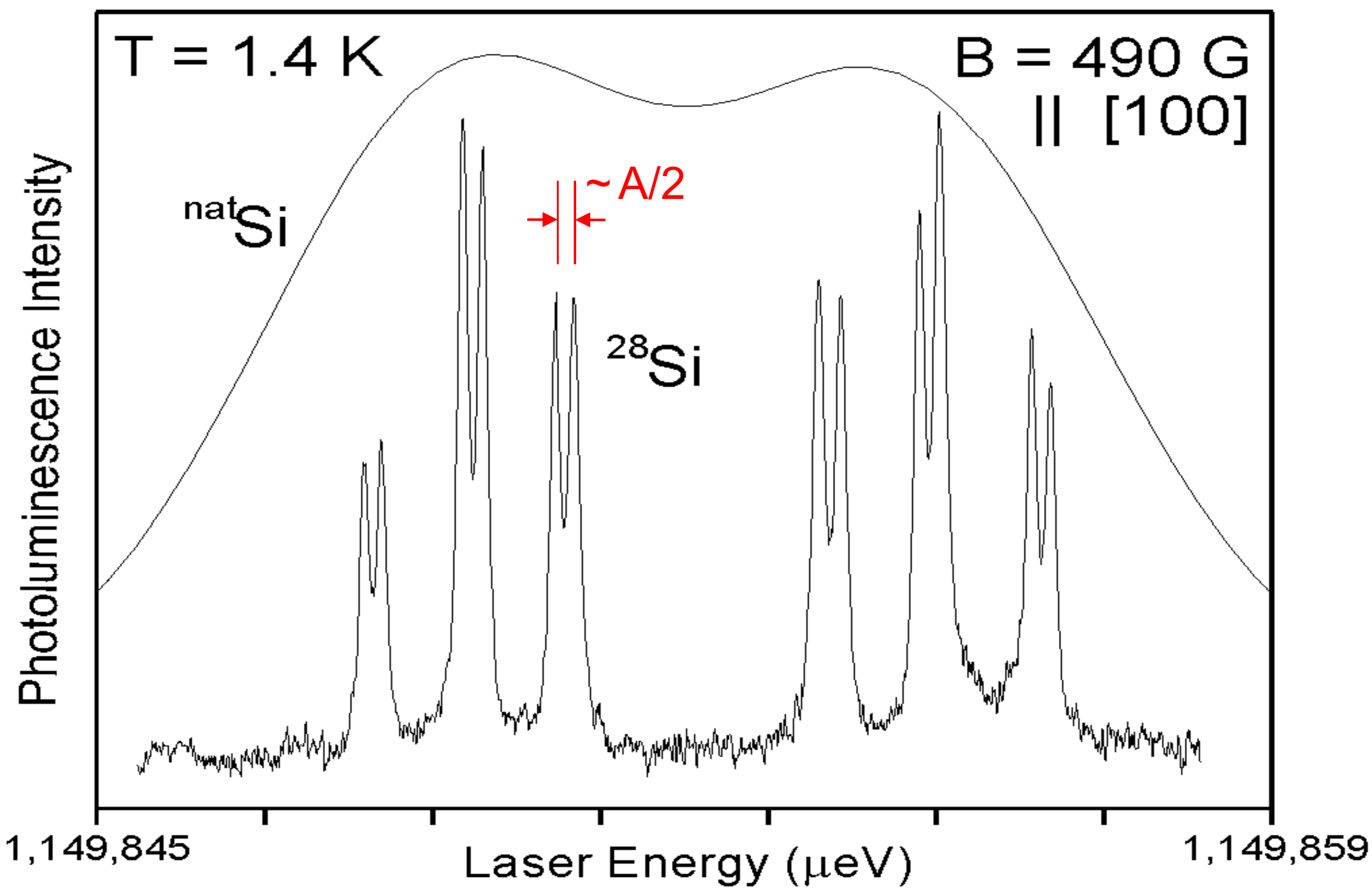
^{31}P donor in Si

Phys. Rev. Lett. **97**, 227401 (2006).

The hyperfine splitting *A*/2 was resolved by high resolution PLE of ^{28}Si:P.

PLE res.	<	^{28}Si:P FWHM	<	A/2	<	PL res.	<	^{nat}Si:P FWHM
0.004		0.15		0.24		1.8		4.7 (μeV)



Group-V Donors in $^{(nat)}$Si

Intermediate-field hyperfine splitting → A/2; Zero-field splitting → (I+1/2)A

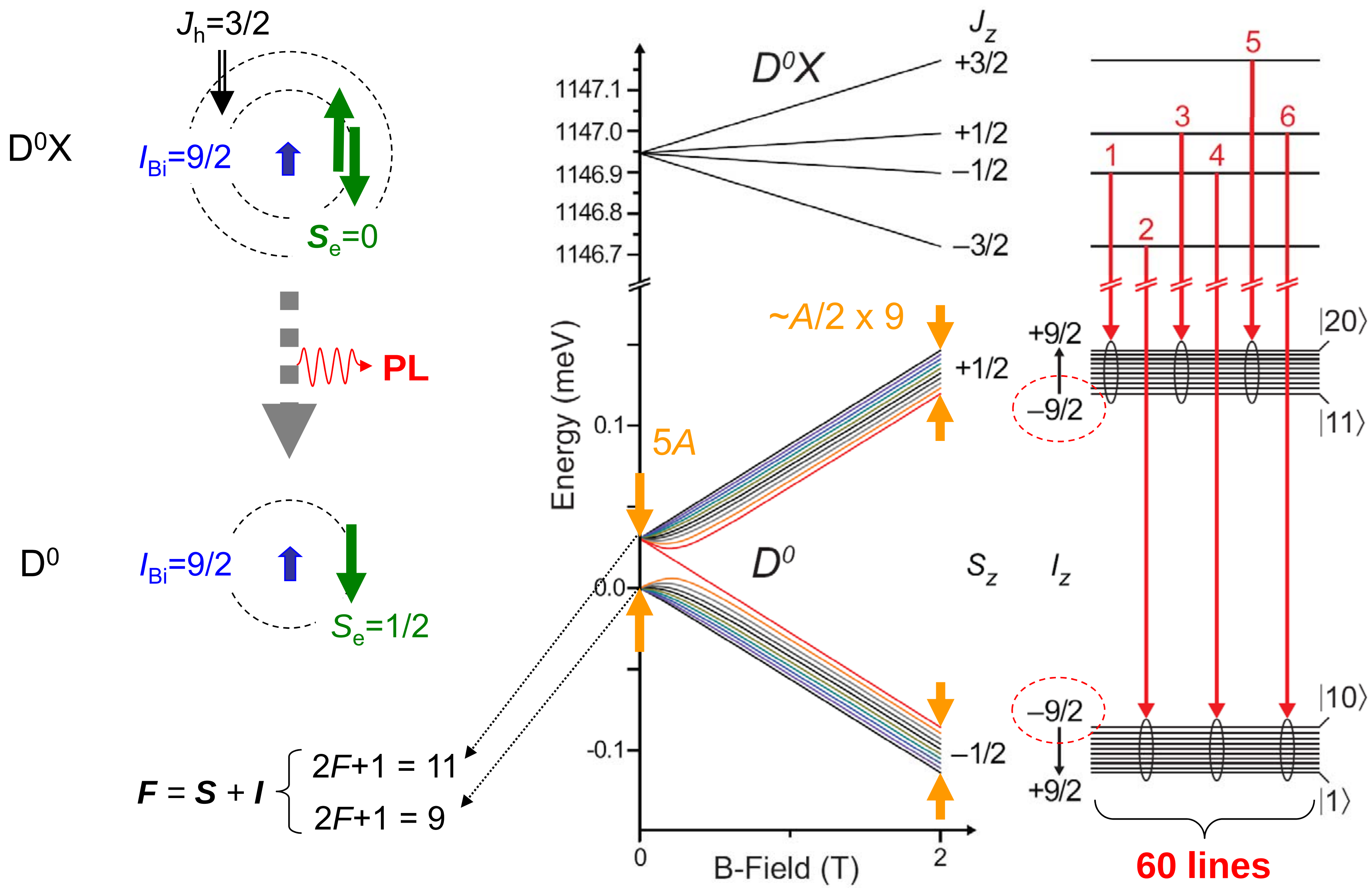
Stable Isotope	E_D (meV)	E_{BE} (meV)	g_e	g_n	I	A/2 (μeV)	(I+1/2)A (μeV)
^{31}P	46	6.17	1.9985	2.2632	1/2	0.243	0.486
^{75}As	54	6.92	1.9984	0.7626	3/2	0.410	1.641
^{121}Sb	43	6.10	1.9986	1.3454	5/2	0.386	2.318
^{123}Sb				0.7285	7/2	0.210	1.679
^{209}Bi	71	9.20	2.0003	0.9135	9/2	3.051	30.51

Feher, Phys.Rev. **114**, 1219 (1959); A.K.Ramdas et al., Rep.Prog.Phys. **44**, 1297 (1981); P. Raghavan, At. Data Nucl. Data Tables **42**, 189 (1989).

- Si:P hyperfine splitting required PLE of enriched ^{28}Si:P.
- Si:Bi hyperfine splitting is larger than PL resolution (1.8 μeV).

Si:Bi BE Energy Diagram

$A/2 = 3.05\ \mu\text{eV},\ g_e\mu_B = 116\ \mu\text{eV/T},\ g_{Bi}\,\mu_n = 29\ \text{neV/T}$

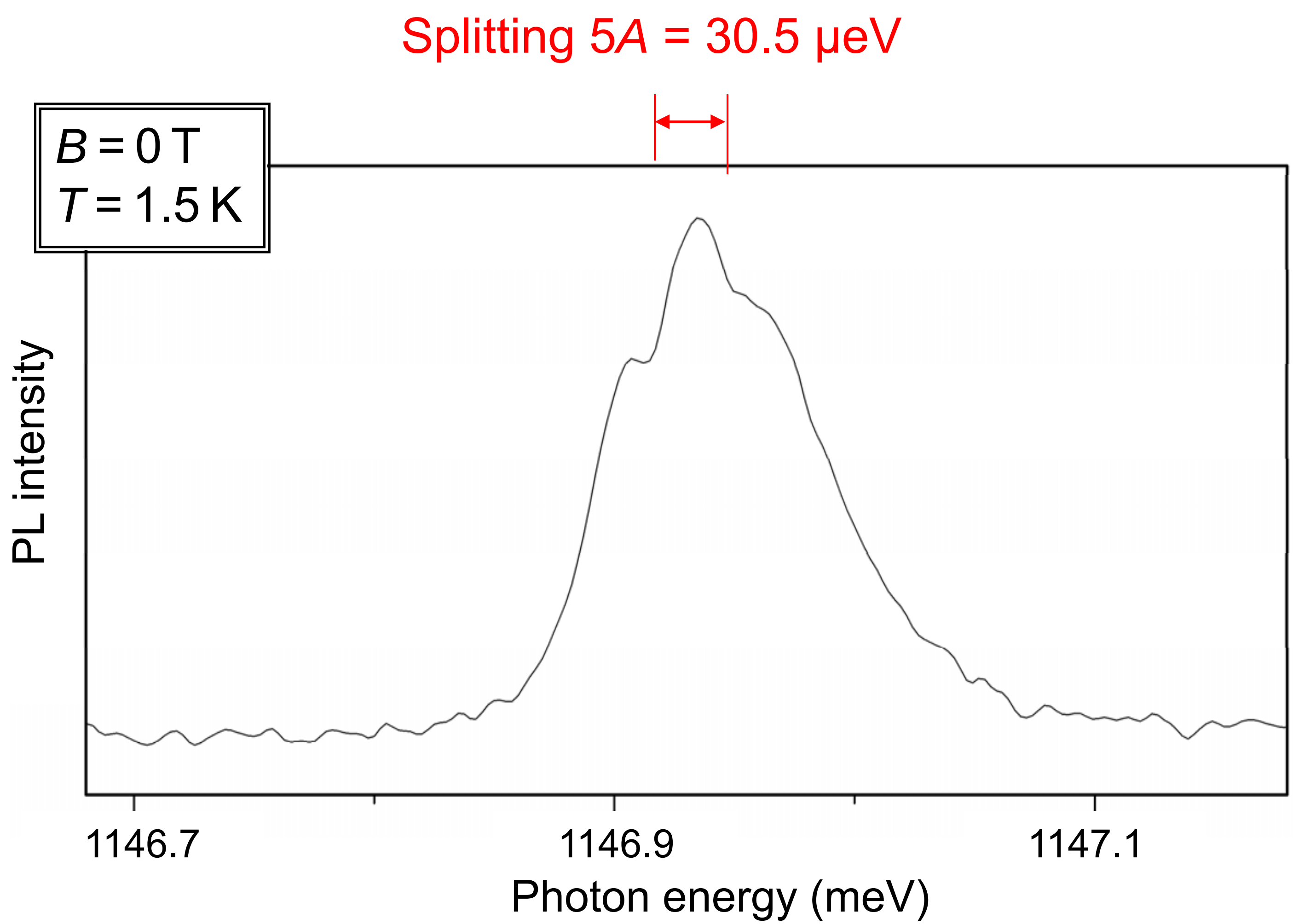


PL Experiments

- ^{nat}Si grown by FZ with Bi doping
  - $\rho = 5.5\ \Omega\cdot$cm, [Bi] $\approx 10^{15}$ cm^{-3}
- Excitation with 1047 nm laser at 400 mW
- No selection of the excitation/emission polarization
- BOMEM DA8 FT spectrometer
  - resolution 1.8 μeV < $A_{Bi}/2 = 3.1$ μeV
- Superconducting magnet
  - B< 7 T; $\mathbf{B}$||[001]; Voigt configuration.
  - inhomogeneity < 0.01%
- 1.5 K $\leq T \leq$ 9 K, in He liquid or gas

Zero-Field Splitting of Bi_{NP} Line

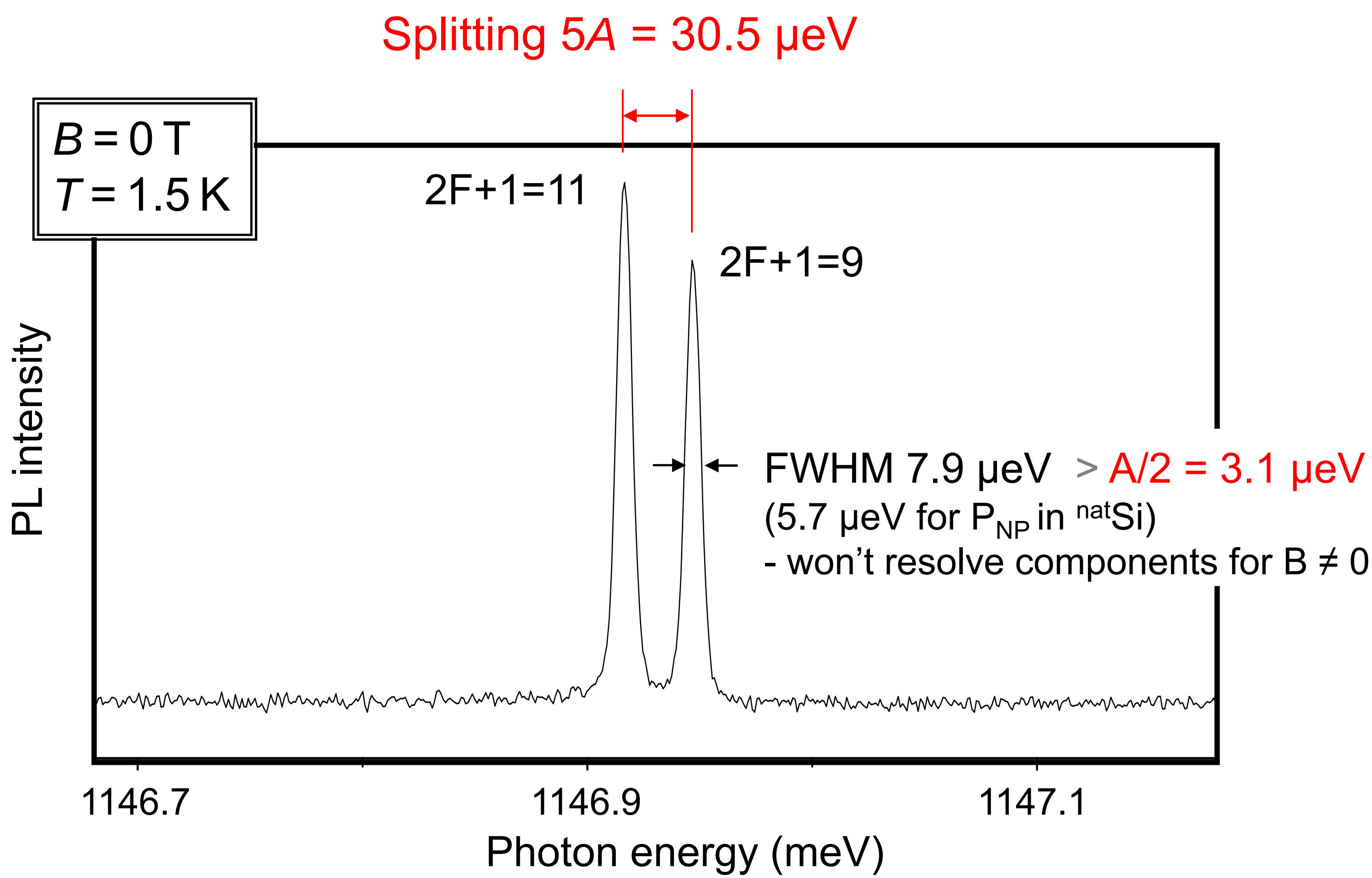
~ 40 year old (Czochralski?) samples won't do…



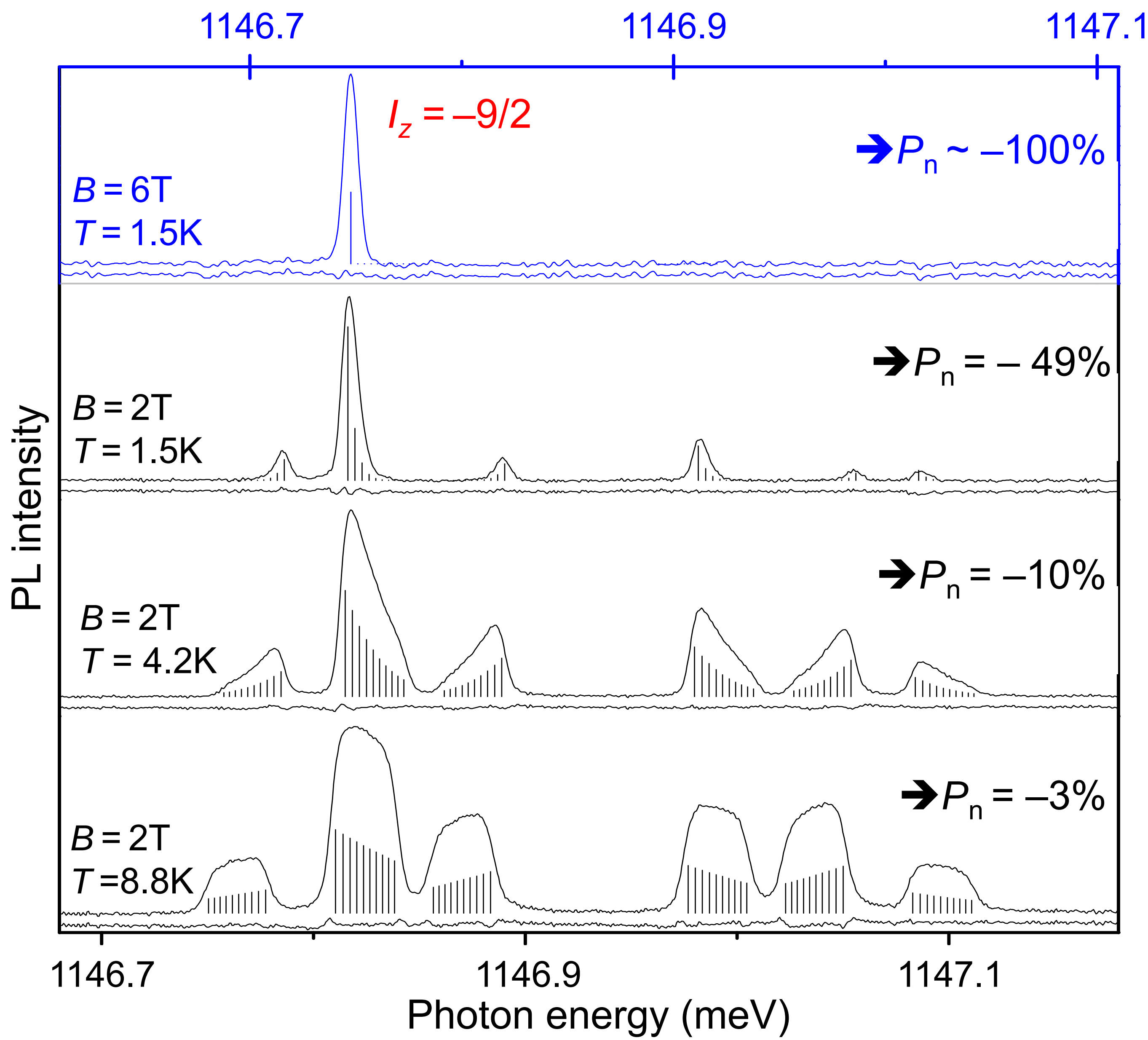
Bi BE first mentioned by Haynes in PRL **4**, 361 (1960)!

Zero-Field Splitting of Bi_{NP} Line

- New floating zone high-purity ^{nat}Si samples:
- Zero-field hyperfine splitting is well-resolved



Bi_{NP} Splitting under Magnetic Field



Model:

- e & h Zeeman splittings
- Hyperfine splitting ~ $A/2$ = 3.05 μeV
- Diamagnetic shift
- Hole thermalization
- Nuclear polarization,

$$P_n \equiv \frac{N(I_z) - N(I_z - 1)}{N(I_z) + N(I_z - 1)}$$

independent of I_z

- $E_g(T)$
- Common line shape and width (~7 μeV)

B and *T* dependent Nuclear Hyperpolarization

This nuclear hyperpolarization seems very similar to the effect for ^{31}P reported by McCamey et al. (PRL **102**, 027601 (2009)) → Overhauser-type model (slow)

B (T)	T_h (K)	$P_e^{(eq)}$ (%)	$P_n^{(eq)}$ (%)	P_n (%)	$N(-9/2)$ (%)
6	1.5	–99	0.07	–79(21)	86(14)
2	1.7	–65	0.02	–54(15)	69(13)
2	4.7	–27	0.007	–10(2)	21(2)
2	8.9	–15	0.004	– 3(1)	13(1)

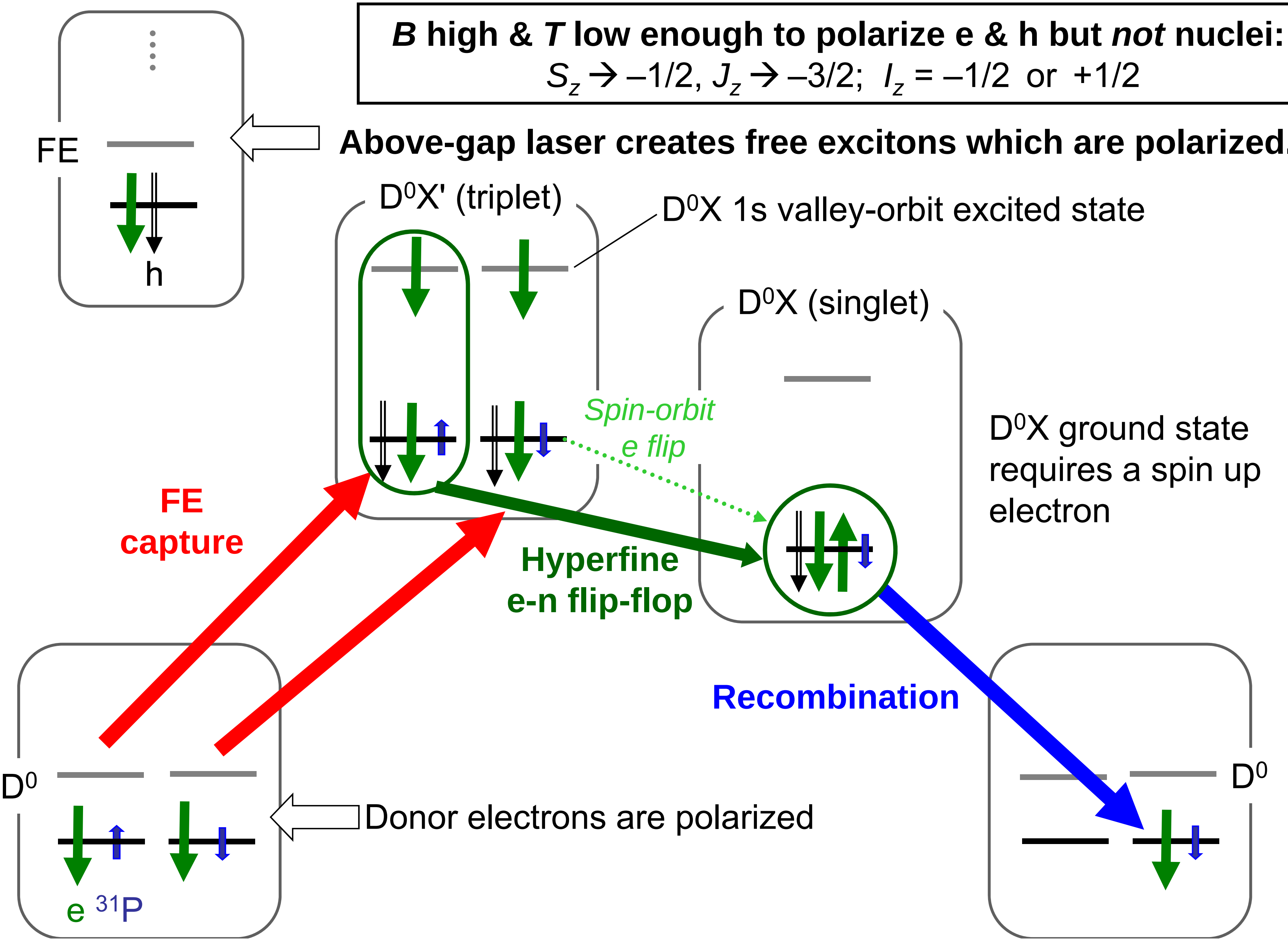
Best fit with 100%

Large uncertainty since individual components cannot be resolved

$$P_n \equiv \frac{N(I_z) - N(I_z - 1)}{N(I_z) + N(I_z - 1)}$$

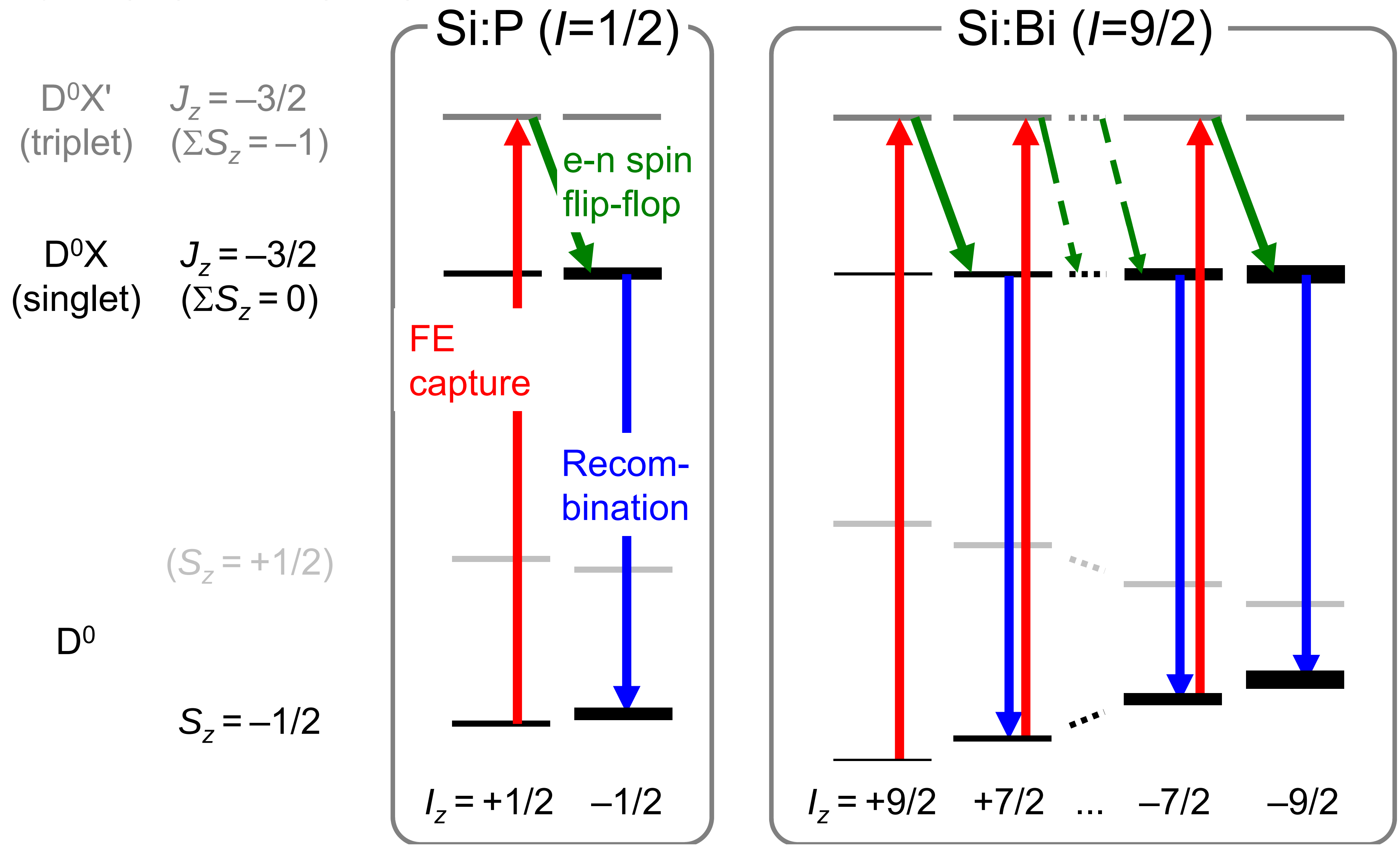
New Nonresonant Optical Hyperpolarization Model (^{31}P)

***B* high & *T* low enough to polarize e & h but *not* nuclei:**
$S_z \rightarrow -1/2$, $J_z \rightarrow -3/2$; $I_z = -1/2$ or $+1/2$

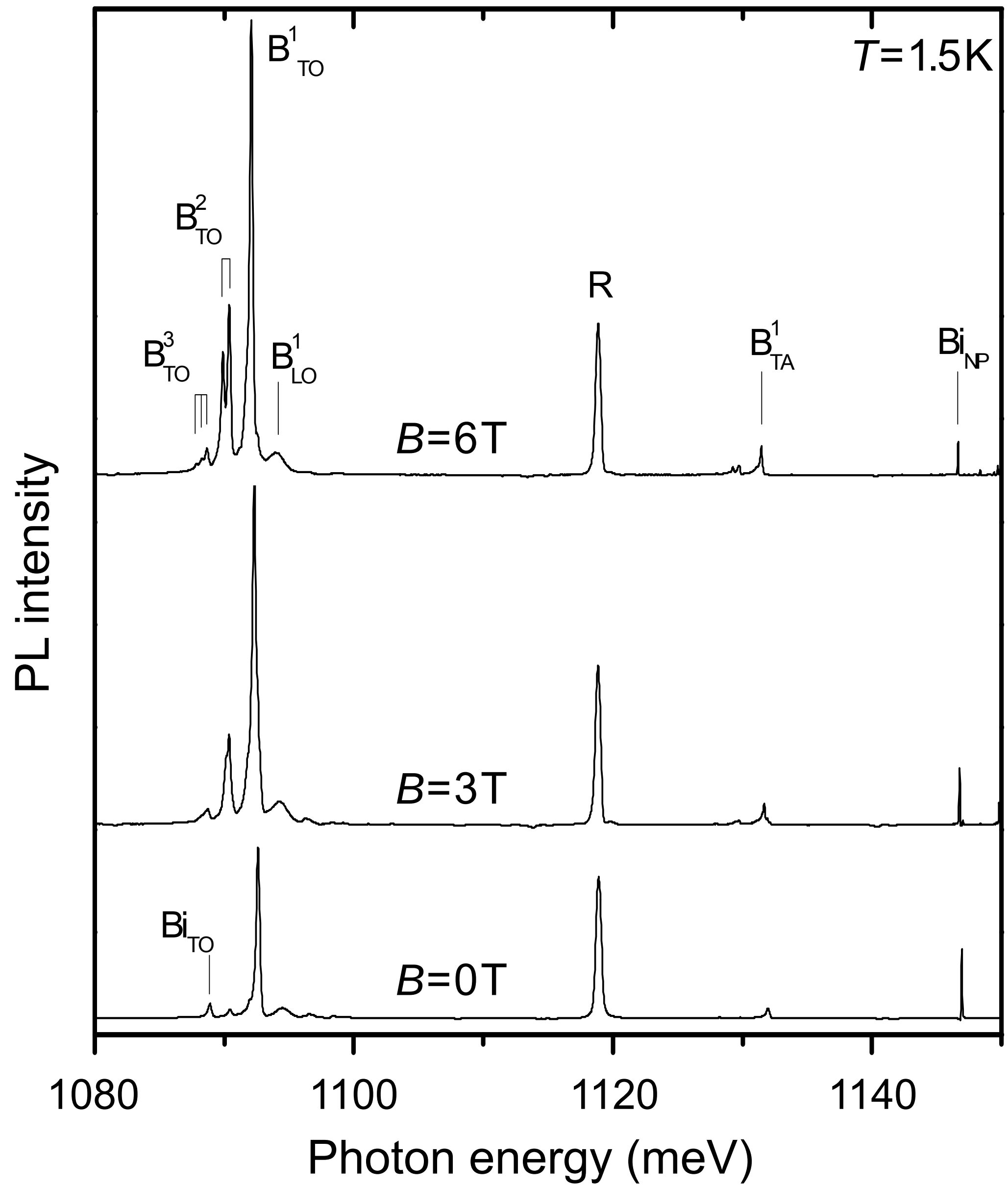


Nonresonant Optical Hyperpolarization of Bi Donor Nuclear Spins

High B (6T), Low T (1.5K)



Support for this new Model



With increasing ***B*** at low ***T***,

– for Bi, the $I_z = -9/2$ state is populated by the optical hyperpolarization;

→ At high *B*, Bi can no longer capture the polarized FE by e-n flip-flop;

→ the Bi PL becomes weaker;

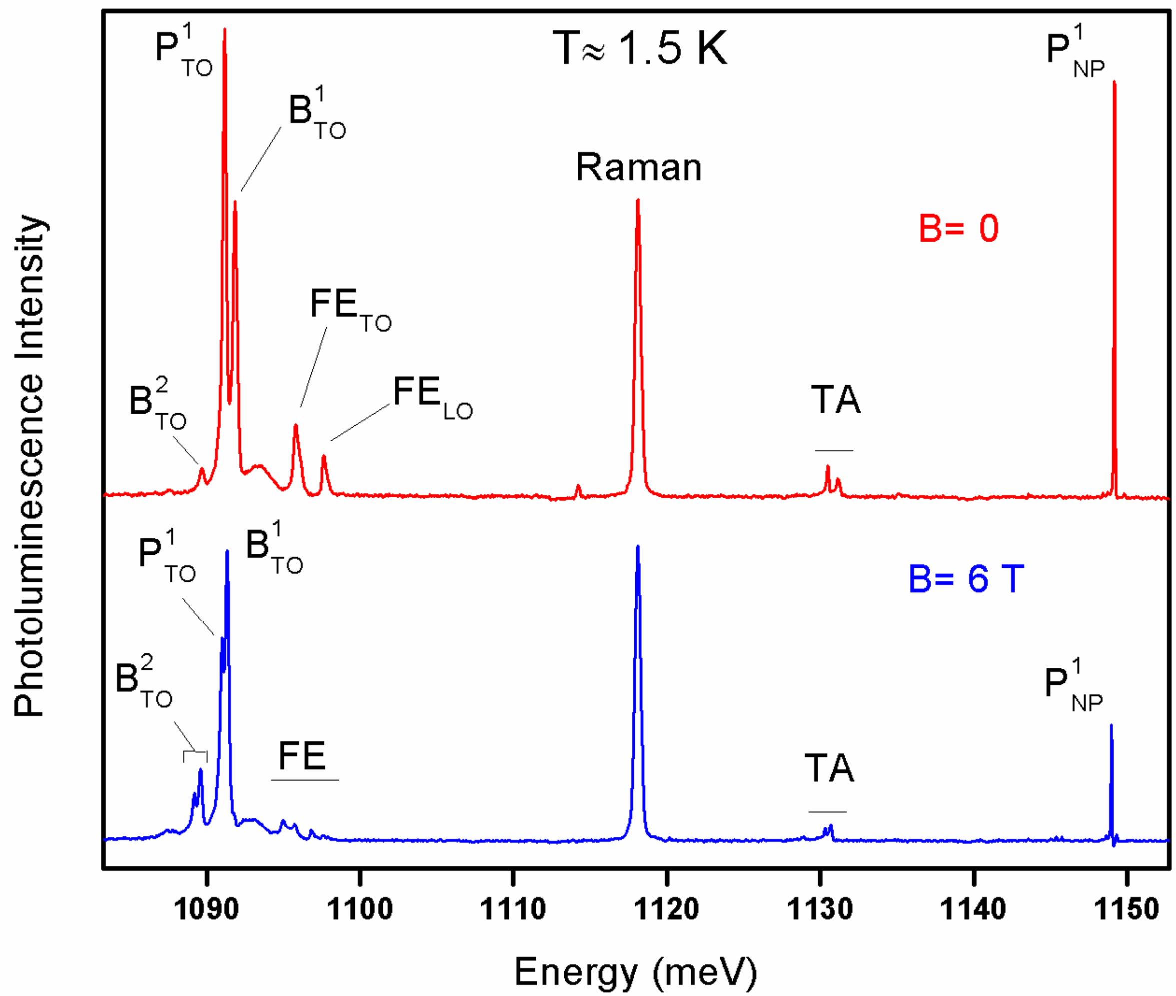
→ the FE density is increased;

– acceptor B can still capture the polarized FE;

→ B PL becomes stronger, especially BMEC (B^2, B^3).

Similar results for ^{31}P

- at high B / low T, P BE becomes weaker, since P polarized to *I* = -1/2 cannot capture polarized FE by the flip-flop process
- FE concentration increases, causing increase in boron PL



Conclusions and future work

- The zero-field hyperfine splitting (30.5 μeV) of the Bi donor in ^{nat}Si is resolved in the PL of D^0X NP line.
- ^{209}Bi nuclear spin is hyperpolarized ($I_z = -9/2$) to nearly 100% at 6T and 1.5K by nonresonant optical excitation.
- We propose a new model for the nonresonant optical hyperpolarization, supported by the PL intensity changes of the donor (Bi, P) vs. acceptor (B) lines with magnetic field.

- The hyperpolariztion may be very fast (sub-ms).
- By employing **^{28}Si:Bi**, all 60 hyperfine components should be resolved even in PL, and the population of each nuclear spin state should be directly measured by PLE or absorption spectroscopy.