Identification and characterization of solid state single photon emitters by means of ab-initio methods

Adam Gali[a,c]

Students:

Viktor Ivády[a,b]
Hugo Pinto[a]
Márton Vörös[c]

Acknowledgments

Jeronimo Maze
Igor Abrikosov

Knut och Alice Wallenbergs Stiftelse
SEVENTH FRAMEWORK PROGRAMME
DIAMANT

Wigner 111 Symposium, Budapest (Hungary), November 12, 2013

[a] Wigner Research Centre for Physics, Budapest, Hungary

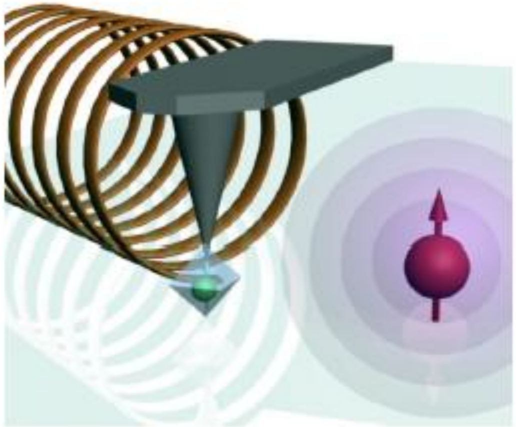
[b] Linköping University, Linköping, Sweden

[c] Budapest University of Technology and Economics, Budapest, Hungary

Outline

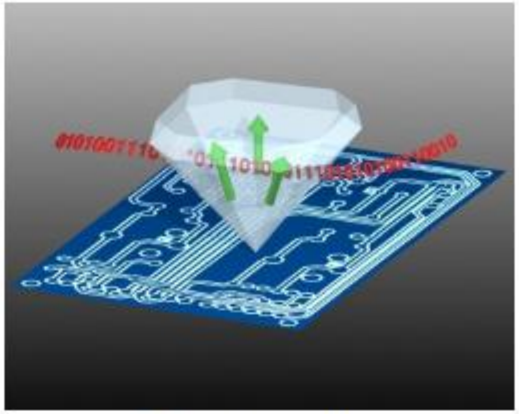
1. Motivation
2. Experimental results of the qubits and single photon sources in diamond and 4H-SiC
3. Theoretical approach for deeper understanding
4. Summary of our findings

NV in diamond: qubit and ultrasensitive sensor



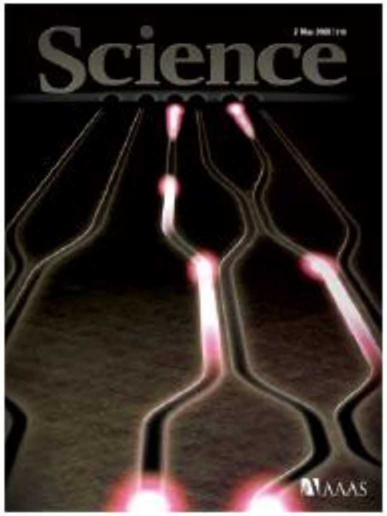
Magnetometry

Nature **455** (2008) pp648–651



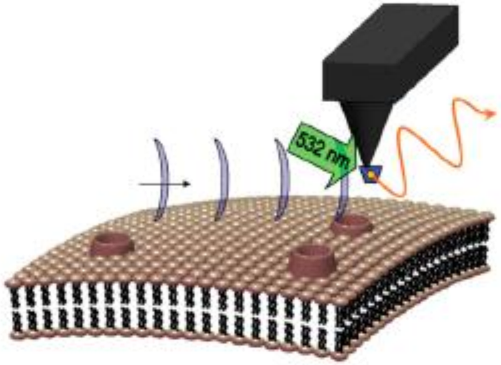
Quantum bit

Science **316** (2007) pp1312–1316



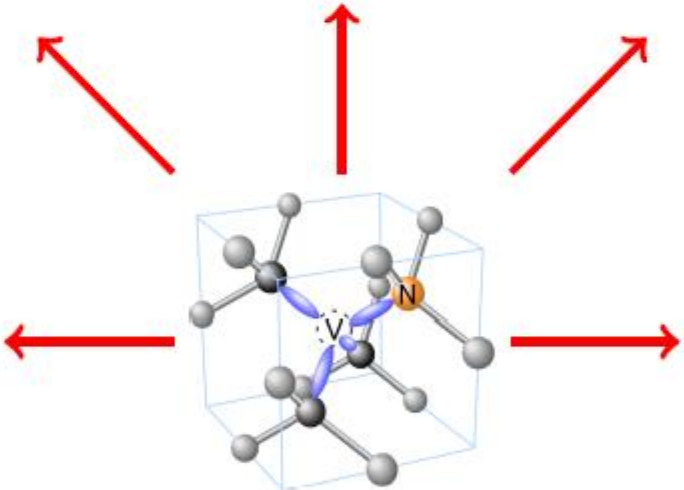
Photonics

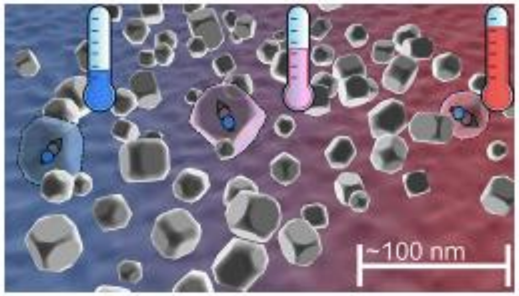
Nature Photonics **5** (2011) pp397–405



Nanosize biomarker

Nature Nano. **6** (2001) pp358–363



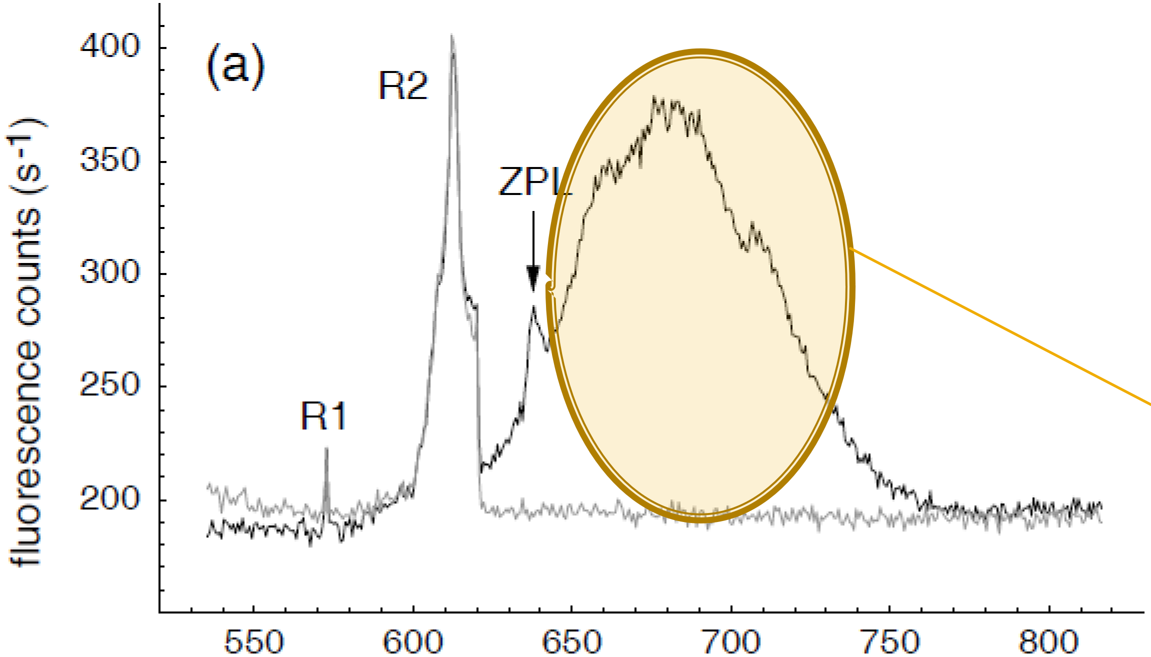


Nanothermometer

PNAS **110** (2013) pp8417–8421

Nano Letters, Nature, Science

SPE in diamond: NV is not perfect



Kurtsiefer et al., PRL **85** 230 (2000)

Phonon Side Band dominates (95%) even at low-T

alternative centers might be useful

Methodology

- *High precision DFT* calculations for converged electronic structures of the point defects
  - Point defects are embedded in large supercells of 500-600 atoms
  - Plane wave bases (E_{cut} = 420eV) and PAW potentials are used as implemented in VASP code
  - Γ-point sampling of the Brillouin-zone
  - Exchange correlation functional: GGA-PBE, hybrid-HSE06
- *Hyperfine*: Fermi-contact + dipol-dipol (+ core polarization)
- We have *implemented* a code to calculate *zero-field splitting* form first principles.
  - The code independent implementation follows the method of Rayson *et. al.*, *Phys. Rev. B* ***77****, 035119 (2008)*

Issue 1:

Split silicon-vacancy center in diamond

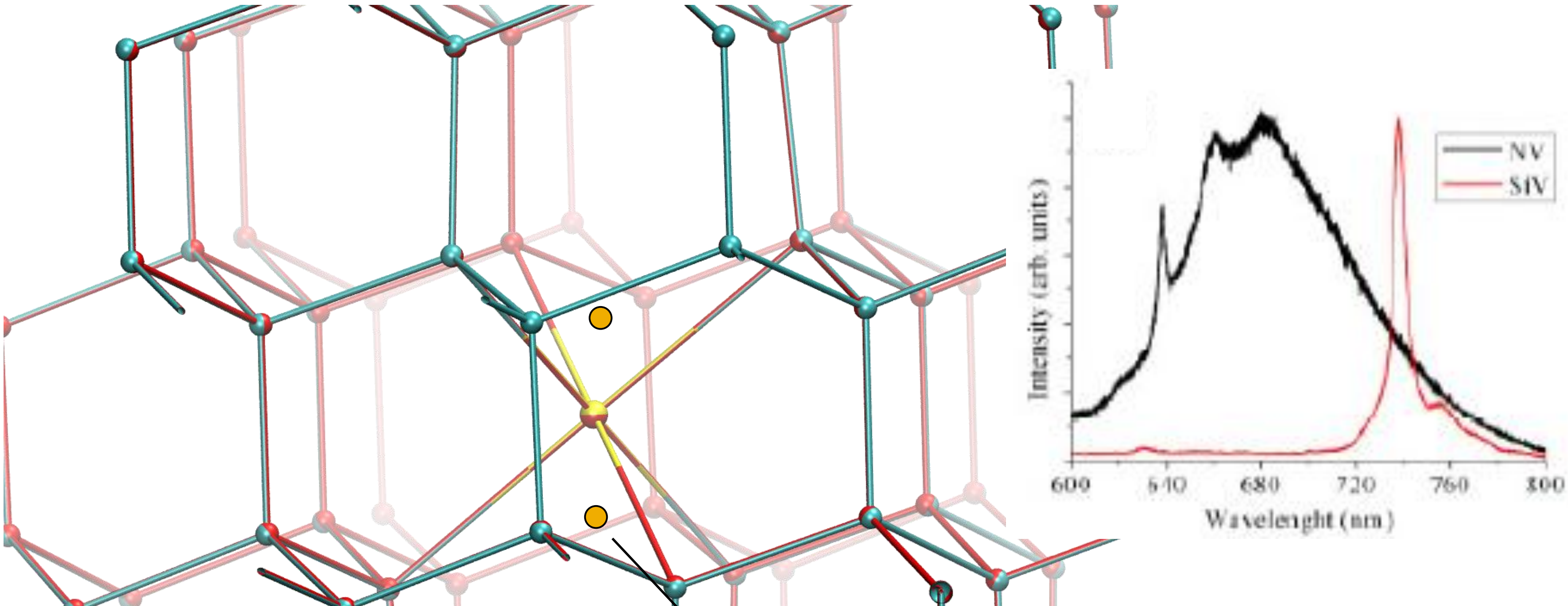
[a] *Wigner Research Centre for Physics, Budapest, Hungary*

[b] *Linköping University, Linköping, Sweden*

[c] *Budapest University of Technology and Economics, Budapest, Hungary*

Bright NIR emitter: SiV defect in diamond

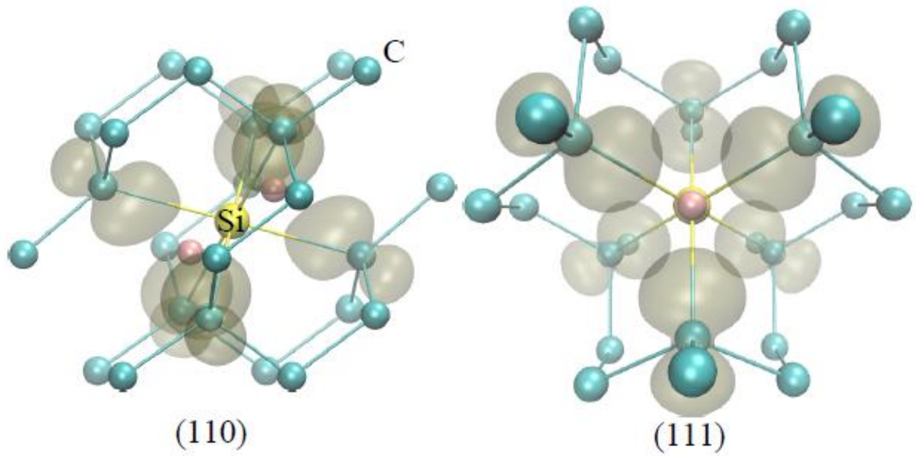
- 1.68-eV PL center as a single source [Wang *et al*., At. Mol. Opt. **39** 37]
- 1.68-eV PL center ↔ *negatively charged* SiV defect [Goss *et al*., PRL **77** 3041]
- 1.31-eV PL center ↔ *neutral* SiV defect [D'Haenens-Johansson *et al*., PRB **82** 155205]

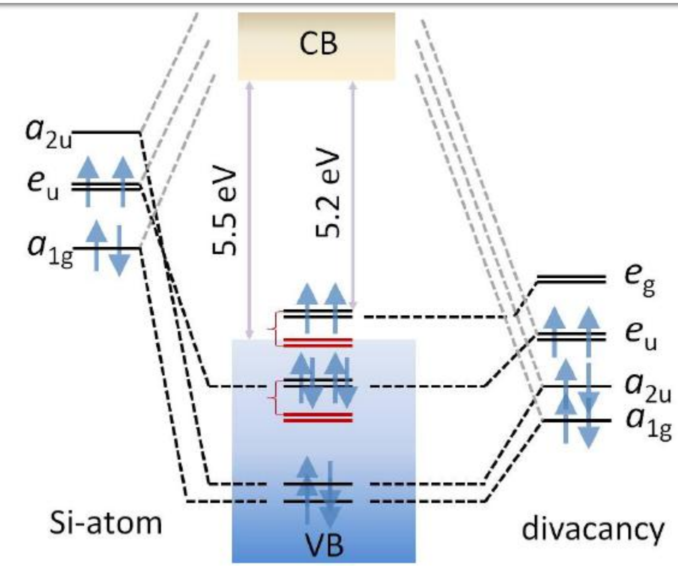


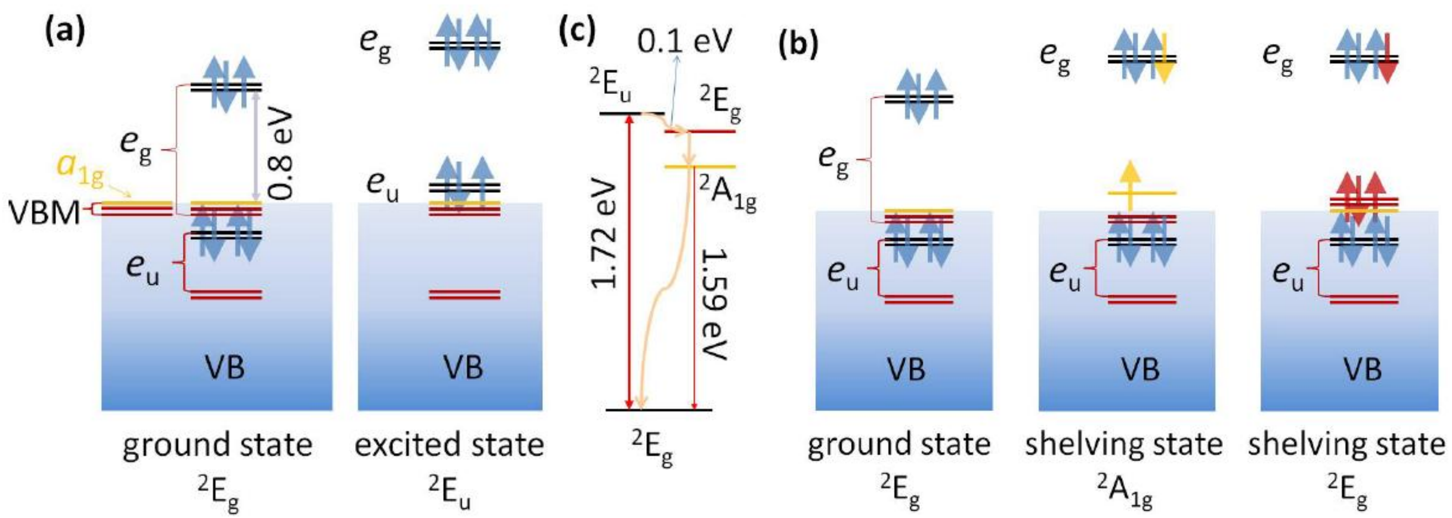
place of C-vacancies

Group theory analysis: D_{3d} symmetry; a_{1g}, a_{2u}, e_u, e_g orbitals

SiV defect in diamond: electronic structure

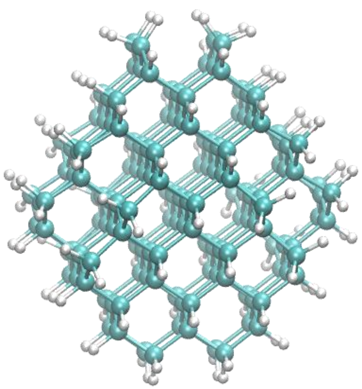




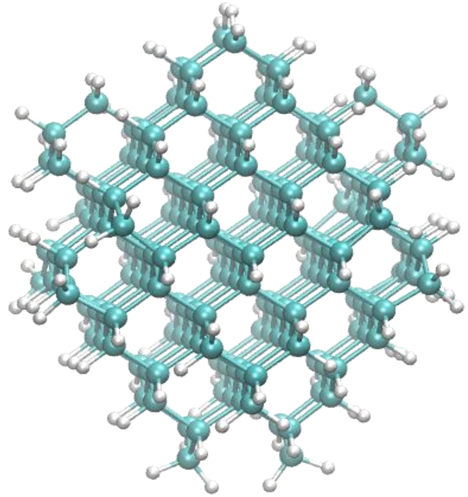


Hydrogenated nanodiamonds

largest quantum confinement limit

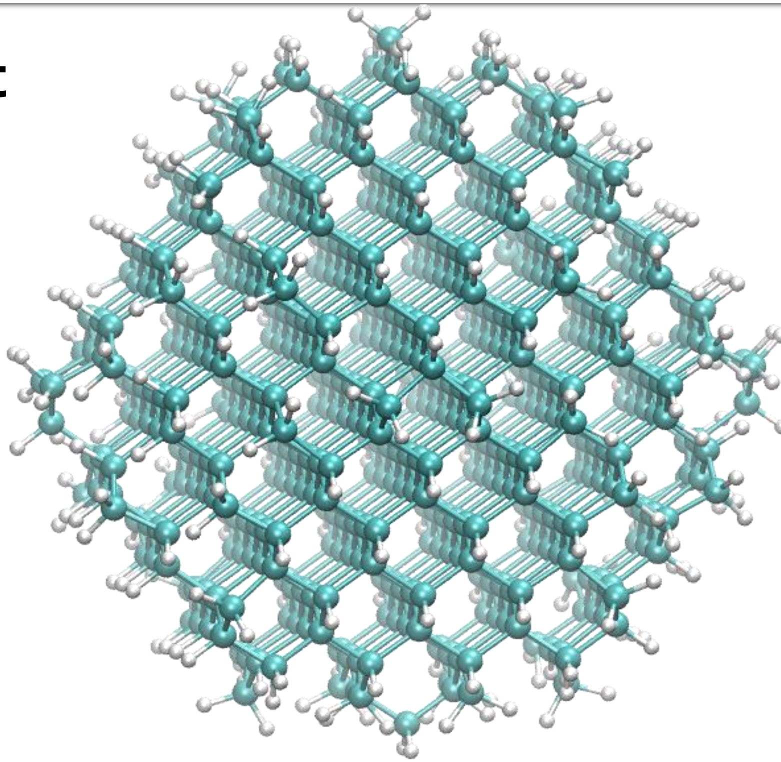


1.1nm



1.3nm

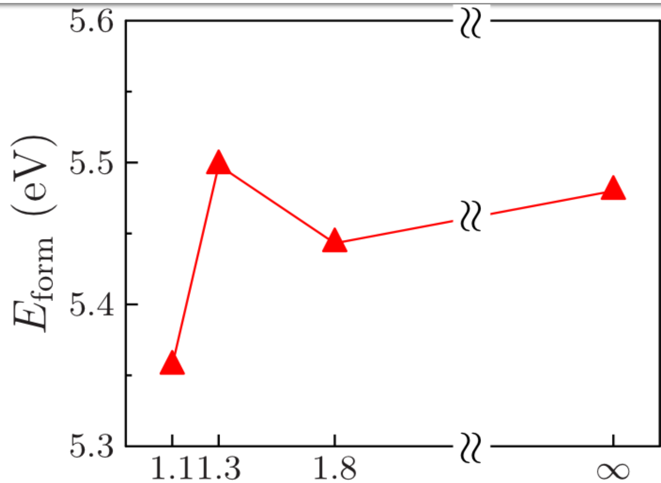
TD-DFT absorption *and* ZPL



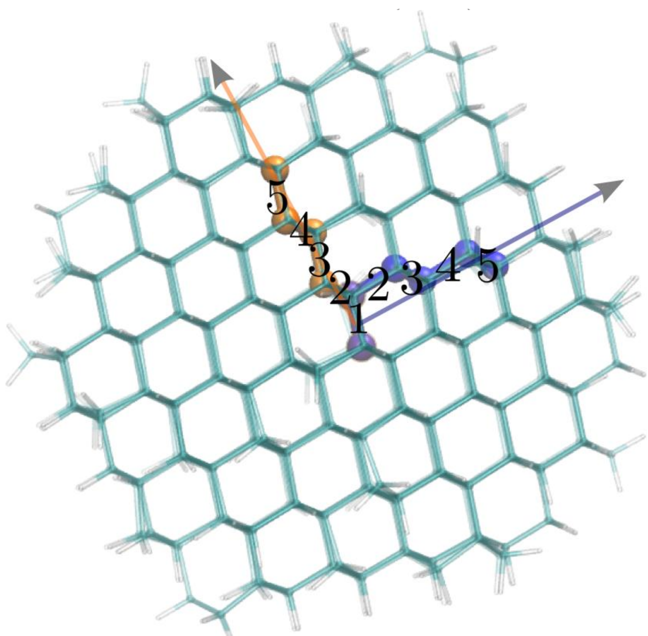
1.8nm

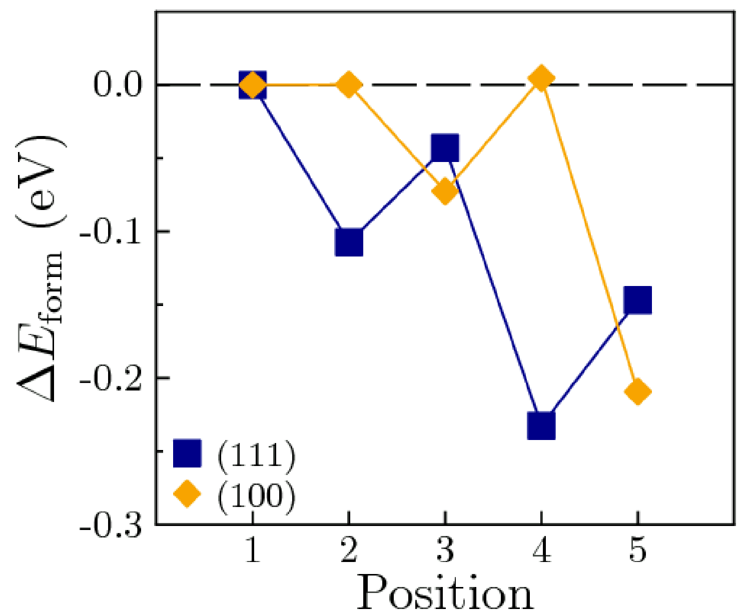
TD-DFT absorption
ZPL is extrapolated from that of 1.3nm

SiV defect in hydrogenated nanodiamonds



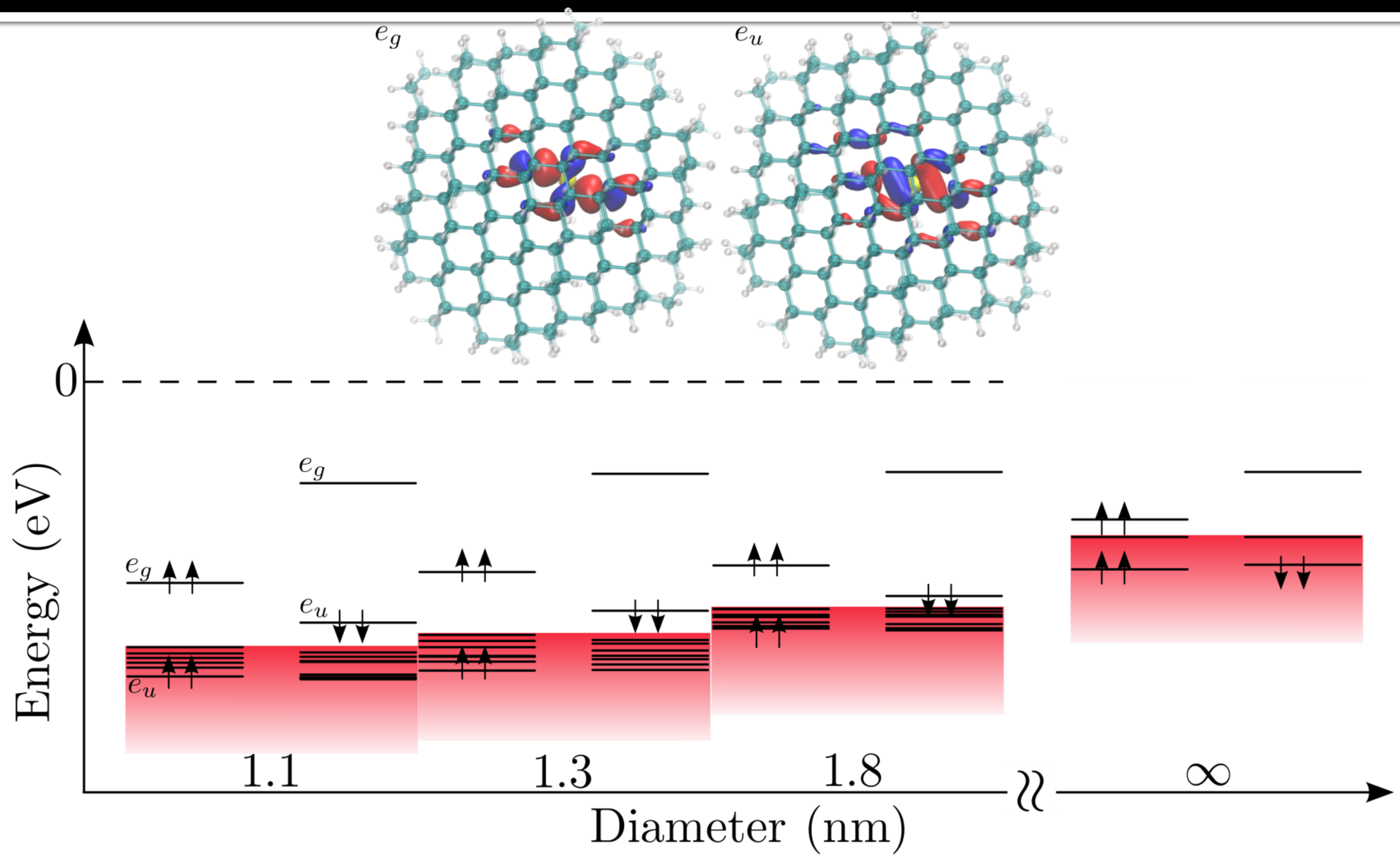
stable even in ultrasmall nanodiamonds





SiV defect in hydrogenated nanodiamonds:

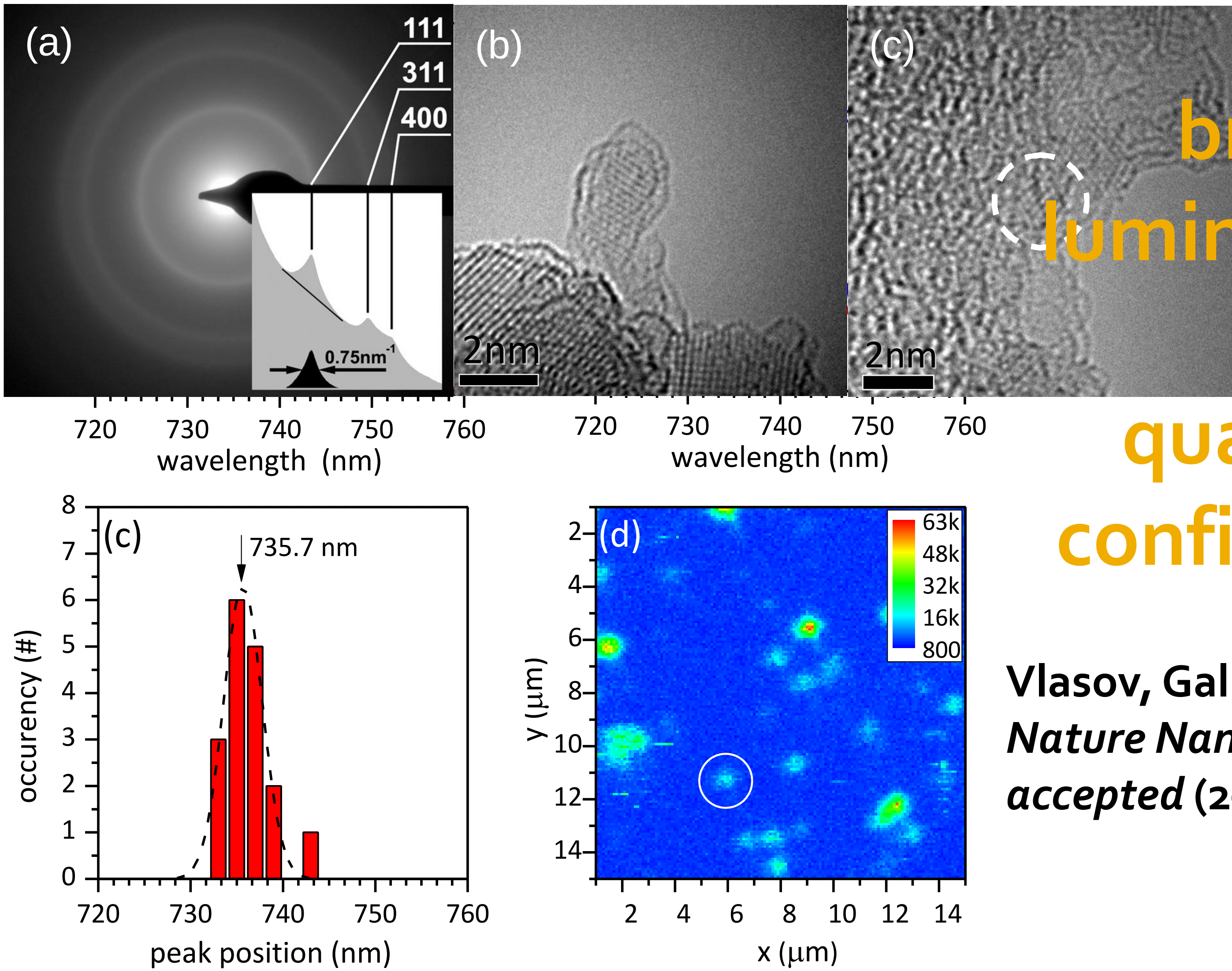
quantum confinement



SiV(–) ZPL: 1.85 eV 1.82 eV 1.78 eV … 1.68 eV

SiV in nanodiamonds: a gift from universe

Efremovka (CV3) and Orgueil (CI) meteorites



bright luminescence & quantum confinement

Vlasov, Gali, Wrachtrup *et al.*, *Nature Nanotechnology*, *accepted* (2013)

Issue 2:

Carbon-antisite vacancy pair in 4H silicon carbide

[a] Wigner Research Centre for Physics, Budapest, Hungary

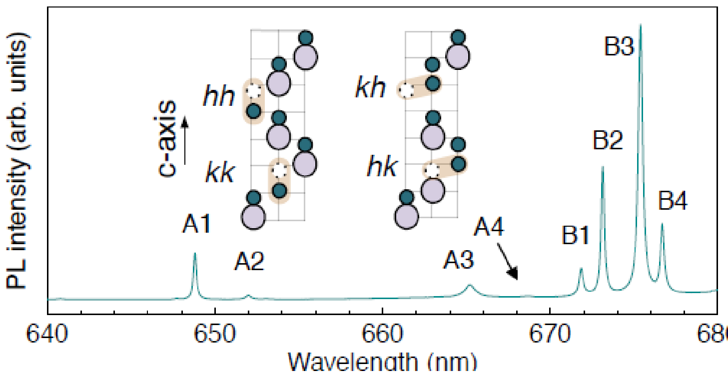
[b] Linköping University, Linköping, Sweden

[c] Budapest University of Technology and Economics, Budapest, Hungary

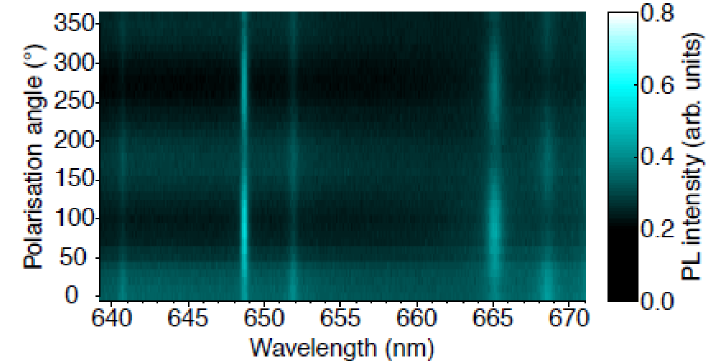
SPE in SiC: Experimental results I.

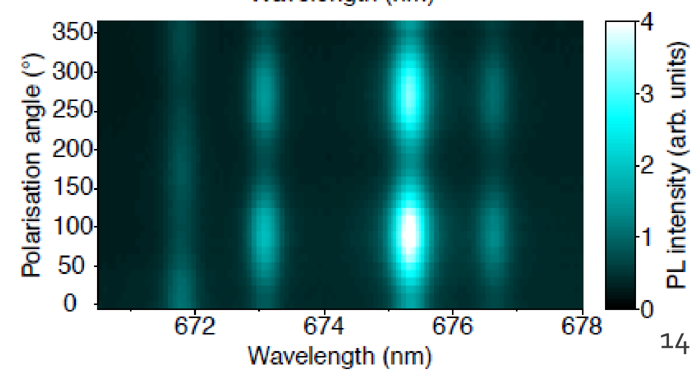
Castelletto, Johnson, Stavrias, Umeda, Ohshima

- 8 observed AB lines
  - 6 AB lines found and associated with $C_{Si}V_C$ [Steeds, PRB **80** 245202 (2009)]



- Emission polarization:
  - A_1 and A_3 are ***E*** || ***c*** polarized
  - A_2 and A_4 are dominantly ***E*** ⊥ ***c*** polarized
  - B_2, B_3 and B_4 are ***E*** ⊥ ***c*** polarized
  - Only B_1 shows ***E*** || ***c***

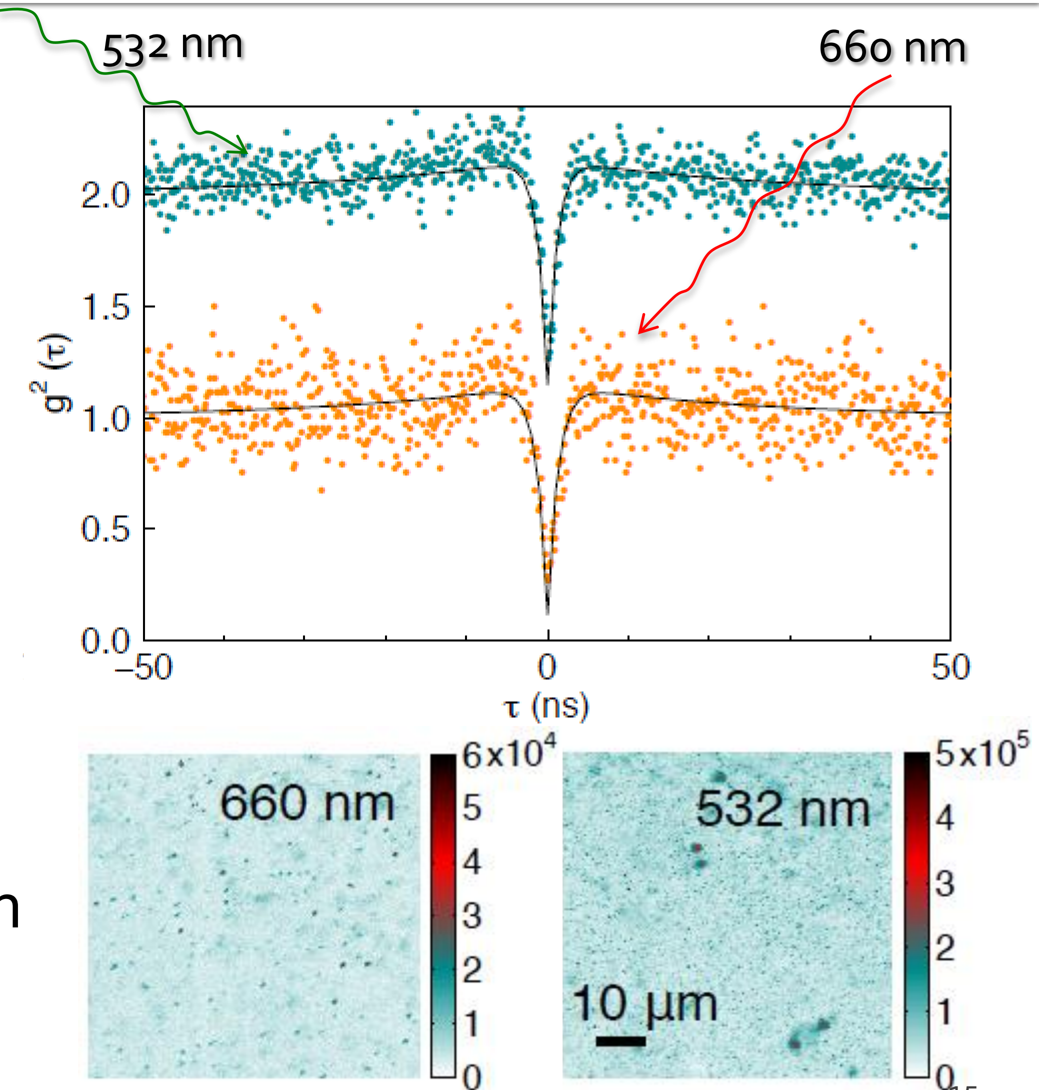




SPE in SiC: Experimental results II.

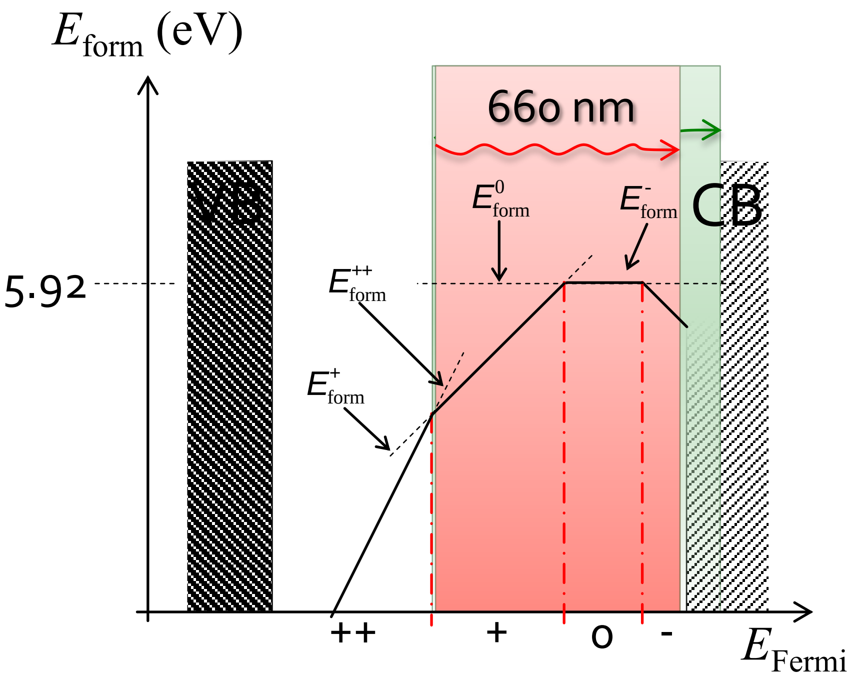
Castelletto, Johnson, Stavrias, Umeda, Ohshima

- Single defect can be isolated
- Excited state lifetime: 1.2 ns
- Quantum efficiency: 70%
- Extreme bright: max 2×10^6 count/s
- Of them 45% blinks
- More blinks at 532-nm excitation than at 660-nm excitation



SPE in SiC: First principles results

Charge transition level (CTL) diagram of $C_{Si}V_C$(hh)



Charge transition levels:

- (++/+): CBM-1.91 eV
- (+/0): CBM-0.92 eV
- (0/-): CBM-0.34 eV

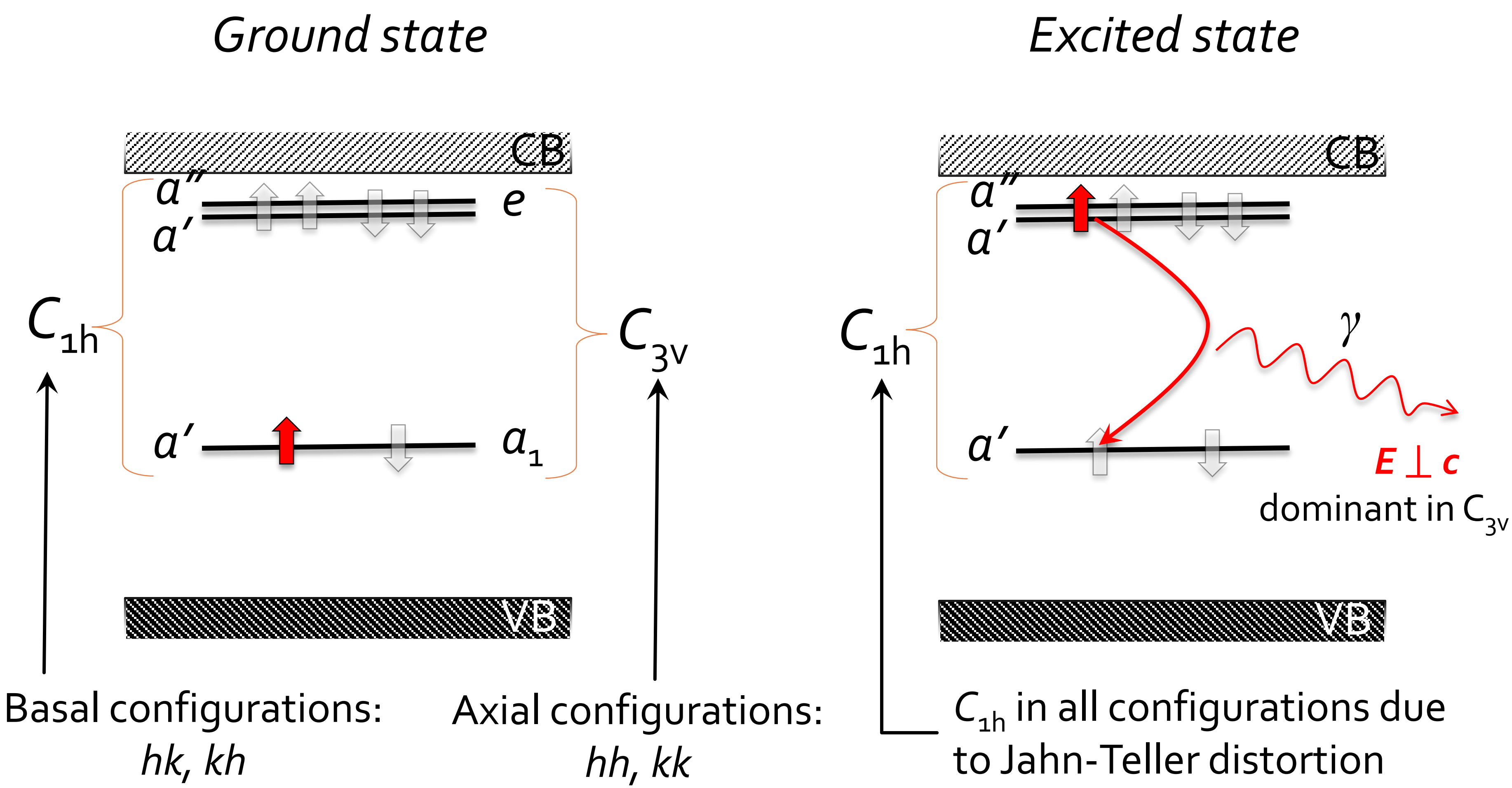
- In $(C_{Si}V_C)^0$: ~ 1 eV is enough to excite an electron to CB
- **No ZPL at ~1.9 eV (678-648 nm) from $(C_{Si}V_C)^0$**
- In $(C_{Si}V_C)^+$: (++/+) CTL:

hh	*hk*	*kk*	*kh*
-1.91 A2	-1.85 A4	-1.83 B2	-1.82 B4

- **Red ZPL is feasible from $(C_{Si}V_C)^+$**
- 2.35-eV vs. 1.88-eV
  1.88eV is more photo-stable

Method: HSE06-DFT, 576-atom supercell, Γ-point
PAW-potentials with 420 eV plane wave cutoff

SPE in SiC: $(C_{Si}V_C)^+$ as model



Group theory considerations I.

- 8 observed AB lines =

  = 2×2 from Jahn-Teller distorted axial configurations +

  + 2×2 from ab ovo C_{1h} configurations

Axial

E splits to $^2A'$ or $^2A''$

$A_1 \rightarrow {}^2A'$	$A_1 \rightarrow {}^2A''$
$\boldsymbol{E} \parallel \boldsymbol{c}$ or $\boldsymbol{E} \perp \boldsymbol{c}$	$\boldsymbol{E} \perp \boldsymbol{c}$
$^2A'$(hh)	$^2A''$(kk)

Basal

Lower excited state can either $^2A'$ or $^2A''$

$\boldsymbol{E} \parallel \boldsymbol{c}$ and $\boldsymbol{E} \perp \boldsymbol{c}$

$^2A'$(hk), $^2A'$(kh)

$C_{1h} \rightarrow$ transversal spin-orbit interaction $\rightarrow$ mix $^2A'$ and $^2A''$ excited states via the ground state

No 100% spin-polarization for $^2A''$ excited state!

Group theory considerations II.

- From the calculated ionization energy + group theory considerations:

$\leftarrow E_{\mathrm{exc}}$

hh		*hk*		*kk*		*kh*	
A''	A'	A''	A'	A'	A''	A''	A'
$\boldsymbol{E} \perp \boldsymbol{c}$	$\boldsymbol{E} \parallel \boldsymbol{c}$	$\boldsymbol{E} \perp \boldsymbol{c}$	$\boldsymbol{E} \parallel \boldsymbol{c}$	$\boldsymbol{E} \parallel \boldsymbol{c}$	$\boldsymbol{E} \perp \boldsymbol{c}$	$\boldsymbol{E} \perp \boldsymbol{c}$	$\boldsymbol{E} \perp \boldsymbol{c}$
A_1	A_2	A_3	A_4	B_1	B_2	B_3	B_4

Agreement

despite A′, no mixture with the ground state wave function

Group theory considerations III.

- In the ground state only 1 electron is in the gap.
- To form $S = 3/2$ shelving state, one electron must be taken from the valence band ($\longrightarrow$ *loosely bound hole)*
- Split VBM edge has a_1 symmetry

	Shelving state	Ground state	Excited state
C_{3v}	^4E	$^2A_1$	^2E
	$a_{1(VB)}^{(1)} a_1^{(1)} e^{(1)}$	$a_{1(VB)}^{(2)} a_1^{(1)} e^{(0)}$	$a_{1(VB)}^{(2)} a_1^{(0)} e^{(1)}$

- Axial spin-orbit may link a_1 orbitals $\longrightarrow$ mix ^4E and ^2E
- Transverse spin-orbit may link e and a_1 orbitals $\longrightarrow$ ^4E and $^2A_1$
- In C_{1h}: states can be mixed in the *basis of* C_{3v} *symm.*
- *In all configuration ^4E can be coupled to $^2A_1$*

Summary on SPE in SiC

- We find that our model can describe the AB PL centers well.
- The dominant polarization both in excitation and radiative emission is perpendicular to the c-axis.
- The occurrence of the metastable shelving state and the process of non-radiative decay via spin-orbit coupling
- The shelving state involves a loosely bound hole near the valance band edge, which may be responsible for the detected large dispersion of lifetimes of the shelving state, as if the hole temporarily leaves the defect then the lifetime of the "dark state" appears longer.
- The nature of the shelving state may also be responsible for the blinking properties.

A Silicon Carbide Room Temperature Single Photon Source, *Nature Materials, accepted*

Thank you for your kind attention!

Contact: Adam Gali, *gali.adam@wigner.mta.hu*