THE BOUND HOLE STATE OF THE NV- CENTER IN DIAMOND

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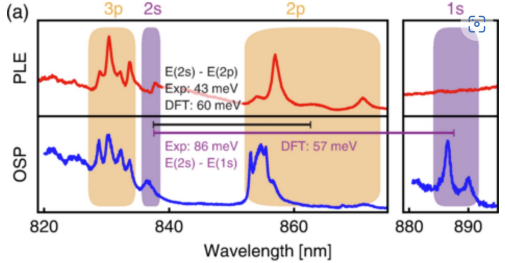
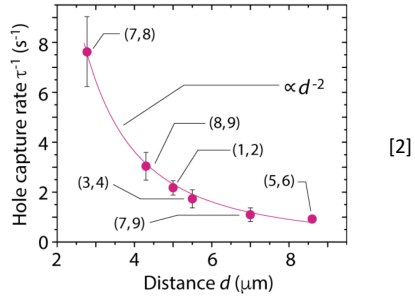
Motivation for studying the bound holes

- Photoionization spectroscopy and possibility of Rydberg physics
  - Recently demonstrated for SiV center
  - Understanding energy levels and their lifetimes are key to observing Rydberg physics
- Generation, transport and capture of holes between NV centers
  - Bound hole states play a critical role in a cascade capture process
  - Lead to a giant capture cross-section
  - Possibly play an important role in NV photocycle



[1]



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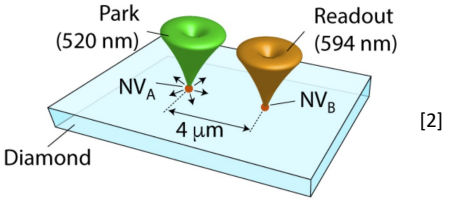

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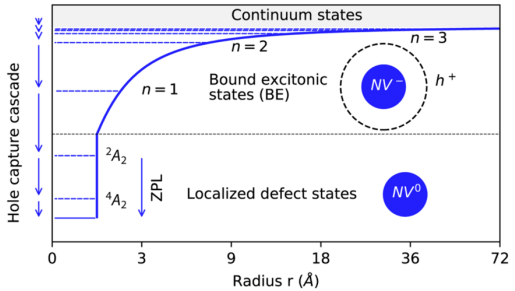

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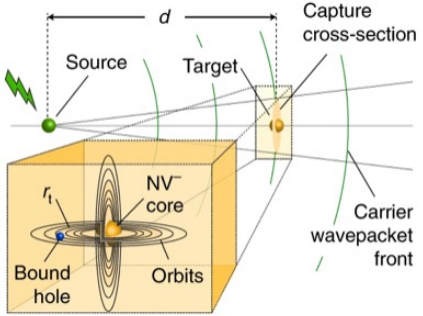


[2]

[1] Z. Zhang et al Phys. Rev. Lett. 125, 237402 (2020).
[2] A. Lozovoi et al Nat. Electronics 4, 717 (2021).

Addressing key issues: characterization of bound states

- No theoretical methods to simulate these bound states and produce important predictions
  - Current approach : density functional theory (DFT), limited by the size of the bound states

- Atomistic details not included in phenomenological models

Key outcomes of this work

- Semi ab initio model of bound states of the NV- center in diamond
  - Theoretical predictions of the bound hole states using effective mass theory
  - Applicable to other deep defects in semiconductors

- Model the non-radiative capture of charge carriers
  - Deformation potential model of acoustic and optical hole-phonon scattering

- First prediction of photoionization spectrum of NV0 to NV- plus bound hole state (**if time permits**)
  - Effective mass theory +DFT
  - Estimation of phonon sidebands

Semi ab initio model of bound hole states

- Adopting an effective mass model
  - Wavefunction ansatz

$$\psi_{b,i}(\vec{r}) = F_{b,i}(\vec{r})u_b(\vec{r})$$

Where $u_b(\vec{r})$ is the periodic Bloch function of the b^{th} band at the valence band maximum (VBM) and $F_{b,i}(\vec{r})$ is the slowly-varying envelope wavefunction to be determined

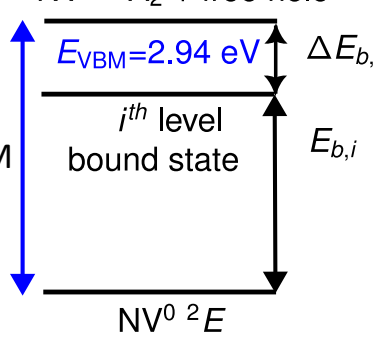
  - Effective Hamiltonian

$$[T_b + \Delta V_b(\vec{r})]F_{b,i}(\vec{r}) = \Delta E_{b,i}F_{b,i}(\vec{r})$$

where $\Delta E_{b,i} = E_{b,i} - E_{VBM}$, E_{VBM} is the free hole energy at the VBM

$$T_b = \vec{p} \cdot \frac{\overleftrightarrow{1}}{2m_b} \cdot \vec{p}$$

$\Delta V_b(\vec{r}) = \frac{1}{\mathcal{V}_c}\int_{\mathcal{V}_c} \eta_b(\vec{r}')\Delta V(\vec{r}' + \vec{r})d^3r'$ is the effective potential for the b^{th} band, $\eta_b(\vec{r}') = |u_b(\vec{r}')|^2$, and $\Delta V(\vec{r}) = V_{NV^-}(\vec{r}) - V_{diam}(\vec{r})$



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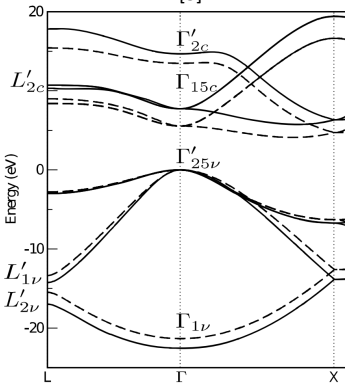


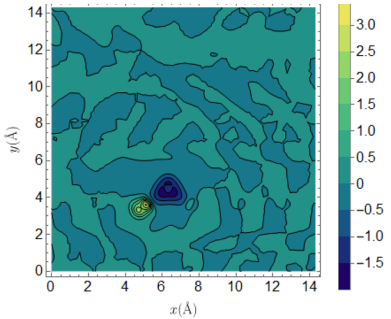
Heavy-hole band
$m_{hh} = 1.08m_e$
Light-hole band
$m_{lh} = 0.36m_e$
[4]

  - Frozen core approximation
    Assume that $\Delta V_b(\vec{r})$ is fixed for all $F_{b,i}(\vec{r})$

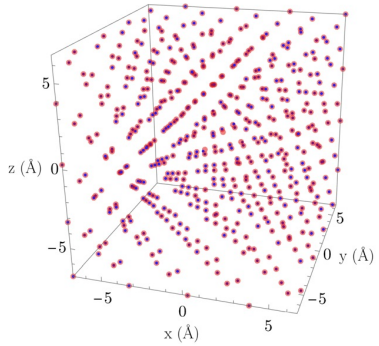
[3] H. Lofas et al AIP Advances 1, 032139 (2011).
[4]O. Madelung, Semiconductors: Data Handbook (Springer, 2004)

Semi ab initio model of bound hole states

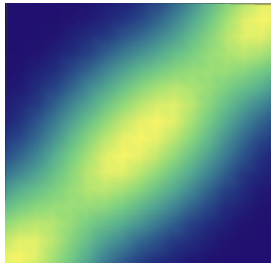
- Construction of the effective potential
  - Employed DFT calculations to obtain the electron densities and nuclear geometries of NV- GS and defect-free diamond
  - Evaluated $\Delta V(\vec{r}) = V_{NV^-}(\vec{r}) - V_{diam}(\vec{r})$
  - Approximated $\eta_b(\vec{r}') \approx \eta(\vec{r}') = \frac{1}{3}\sum_b |u_b(\vec{r}')|^2$
  - Approximated $\eta(\vec{r}')$ by a Gaussian function



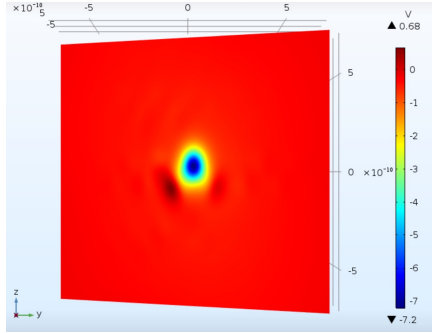
Difference in e^- charge density in $(1\bar{1}0)$ plane



Nuclear geometries



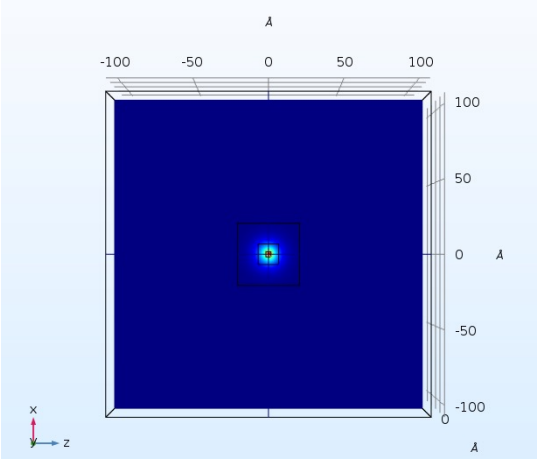
$\eta(\vec{r}')$ in $(1\bar{1}0)$ plane



Constructed $\Delta V_b(\vec{r})$ in $(1\bar{1}0)$ plane

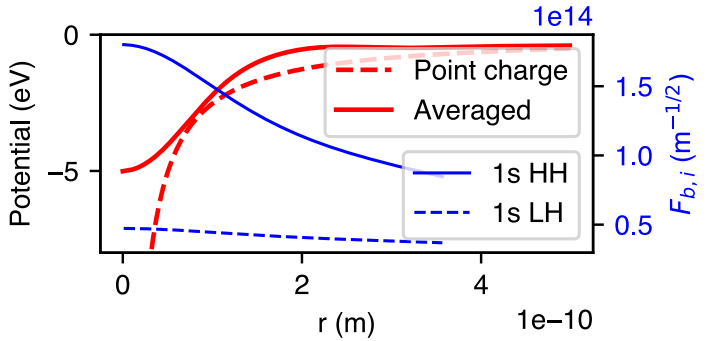
Semi ab initio model of bound hole states

- COMSOL solutions

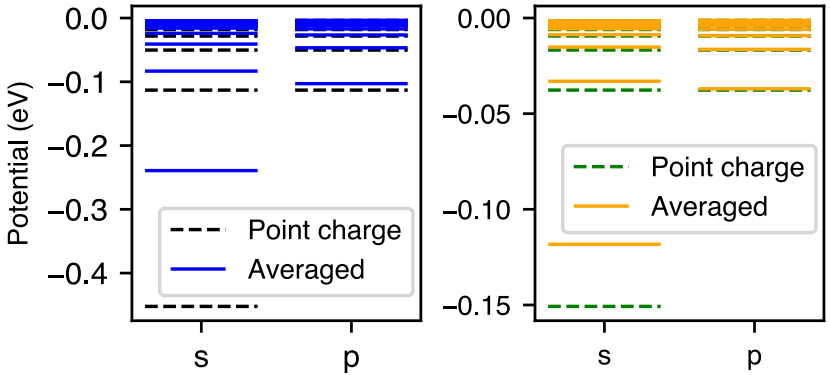


- Lowest energy eigenstate (-0.24 eV) solved in COMSOL
- Ab initio calculations (-0.27 ev) [2]

- Solutions for approximation of spherical symmetry



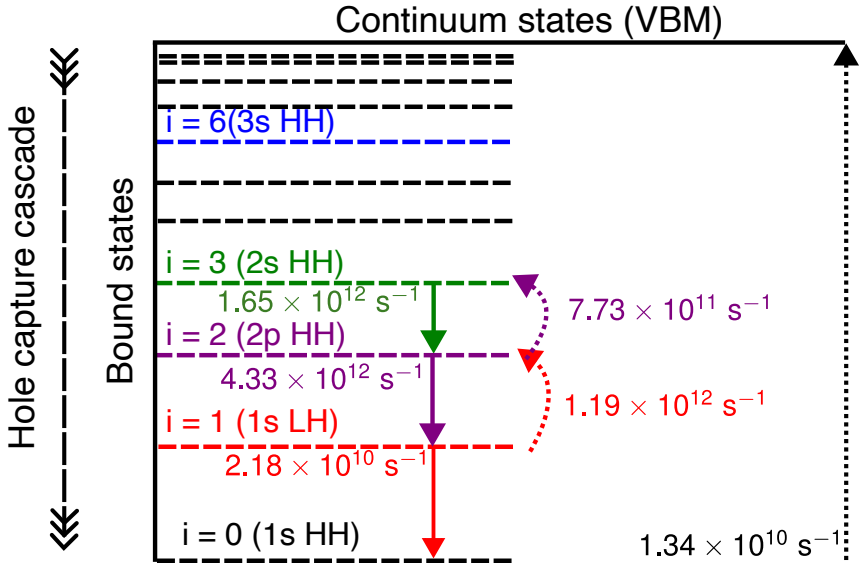
Red : comparison of spherically-averaged $\Delta V_b(\vec{r})$ with a monopole potential in diamond.
Blue : Radial wavefunctions



Comparison of l=0 and l=1 eigenenergies between a point charge and heavy hole (left) and light hole (right)

Non radiative capture of bound holes

- Hole cascade capture
  - Critical transition from the first excited state to the ground state
- Deformation potential model
- Emission rate of first excited state is the hole capture rate
- Assumption of thermal equilibrium (TE)
- $\sigma_{cap} \approx 2\times10^{-4}\mu m^2$
  - 10 times smaller compared to experiments [2]
- Exclusion of second order perturbation
  - One-phonon process dominates at low temperature [5]
  - Two-phonon process scales as T^5 at room temperature [6]



Left (solid lines) : emission rate

Right (dotted lines) : absorption rate. All values are at T=300 K

[5]T. Astner et al, Nature Materials 17, 313 (2018).
[6] M. W. Doherty et al, Physics Reports 528, 1 (2013)

Summary

- Presented a first attempt at a semi ab initio approach to modelling bound hole states which are applicable to other semiconductors

- Modelled the non-radiative capture of charge carriers with a capture cross-sections of $\approx 2\times10^{-4}\mu m^2$

Future work

- Improve the current implementation of the approach
  - Improve model of the probability density
  - Include second order perturbation in the scattering rate calculations
  - Improve model of capture cross-sections to include bound states at low temperatures

THANK YOU

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