Multi-electron multi-valley effective mass theory: Numerical considerations and applications

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Overview

Multi-valley effective mass theory (MV-EMT) is quantitatively accurate for 1-electron properties

- Tunnel coupling between phosphorus donors [1]
- Valley-splitting of quantum dots + disorder [2]

Multi-electron effects are necessary to study

- Donor-dot hybrid systems [3] (see V. Srinivasa's poster)
- Multi-electron quantum dots where spin-orbit [4], valleys, and disorder come into play (see R.M. Jock's talk, N.T. Jacobson's poster)

High accuracy solutions of MV-EMT equations are especially critical for multi-electron effects

Theory

Single-particle orbitals = **envelope** + **Bloch** functions

$$\psi_i(\mathbf{r}) = \sum_{\mu \in \pm\{x,y,z\}} F_{i,\mu}(\mathbf{r}) e^{i\mathbf{k}_\mu \cdot \mathbf{r}} u_\mu(\mathbf{r})$$

Shindo-Nara equations describe **interaction-free** problem

$$\hat{H}_{0,i}\psi_i(\mathbf{r}) = \varepsilon_i^0 \psi_i(\mathbf{r})$$

$$\hat{T}_\mu F_{i,\mu}(\mathbf{r}) + \sum_{\nu \in \pm\{x,y,z\}} V^{VO}_{\mu,\nu}(\mathbf{r}) F_{i,\nu}(\mathbf{r}) = \varepsilon_i^0 F_{i,\mu}(\mathbf{r})$$

For N>1 electrons, the **interacting** Hamiltonian is:

$$\hat{H} = \sum_{i,\sigma} \varepsilon^0_{i,\sigma} \hat{c}^\dagger_{i,\sigma} \hat{c}_{i,\sigma} + \frac{1}{2} \sum_{\substack{i,j,k,l \\ \sigma,\sigma'}} V_{ijkl} \hat{c}^\dagger_{i,\sigma} \hat{c}^\dagger_{j,\sigma'} \hat{c}_{k,\sigma'} \hat{c}_{l,\sigma}$$

Coulomb interaction is **valley-local**

$$V_{ijkl} \approx \sum_{\mu,\nu} \int\int d\mathbf{r} d\mathbf{r}' \frac{F^*_{i,\mu}(\mathbf{r}) F^*_{j,\nu}(\mathbf{r}') F_{k,\nu}(\mathbf{r}') F_{l,\mu}(\mathbf{r})}{4\pi\epsilon_{Si}|\mathbf{r}-\mathbf{r}'|}$$

Move through hierarchy of **electronic structure methods**

- -Slater exchange / semi-local DFT
- -Hartree-Fock
- -Perturbation theory
- -Configuration interaction

Challenges

A lot of "magic" that differentiates dots + donors from conventional quantum chemistry

Numerical issues:

- -**Difficult to systematically improve** basis sets for isolated donors, interfaces, and hybrids thereof
- -Symmetries that facilitate creation of good virtual orbitals for correlation **can no longer be exploited**
- -Quartic scaling costs amortized by **small basis sets**
- -Shindo-Nara requires **non-standard matrix elements**

Physical issues:

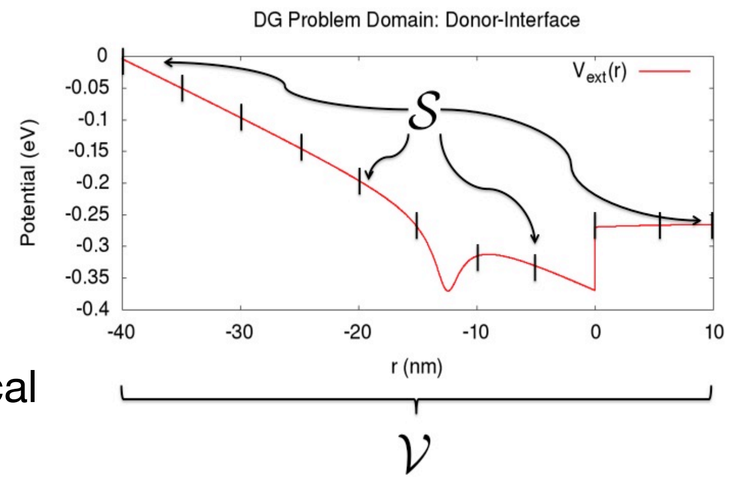
- -Proper treatment of **effective Coulomb interaction?**
  - -**Image charges** due to oxide
  - -**Local field corrections** for short-range physics
- -Modifications to bulk central cell corrections?

Numerical Method

Domain decomposed into hexahedral elements

Element-local basis
-Chebyshev polynomials
-plane waves
-Gaussians

Interior penalty (IP) methods used to glue together high-order local solutions



IP discretization of Shindo-Nara + Hartree-Fock

Find $F_{i,\mu}$ in the span of $u_{j,k}$ that is a stationary point of

[[ ]] = jump {{ }} = average

$$\frac{1}{2}\left(\langle \nabla u_{j,k}, \nabla F_{i,\mu}\rangle_{\mathcal{V}} - \langle \{\{\nabla u_{j,k}\}\}, [[F_{i,\mu}]]\rangle_{\mathcal{S}} - \langle [[u_{j,k}]], \{\{F_{i,\mu}\}\}\rangle_{\mathcal{S}} + \frac{\alpha}{h}\langle [[u_{j,k}]], [[F_{i,\mu}]]\rangle_{\mathcal{S}}\right)$$

Kinetic energy

$$\sum_{\nu \in \pm\{x,y,z\}} \langle u_{j,k}, V^{VO}_{\mu,\nu} F_{i,\nu}\rangle_{\mathcal{V}} + \langle u_{j,k}, V^{HF}_{\mu} F_{i,\mu}\rangle_{\mathcal{V}}$$

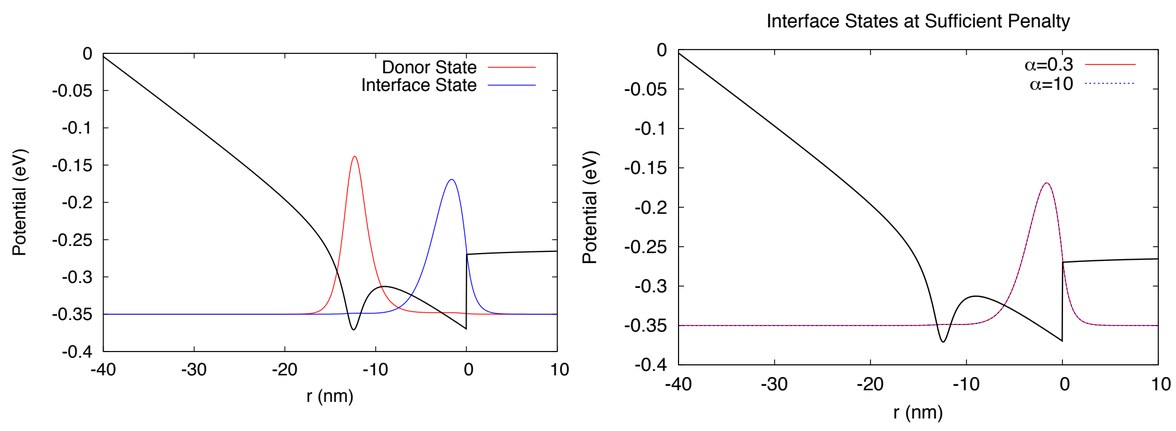
External potential + Hartree-Fock mean field

Mean field computed using IP Poisson solve

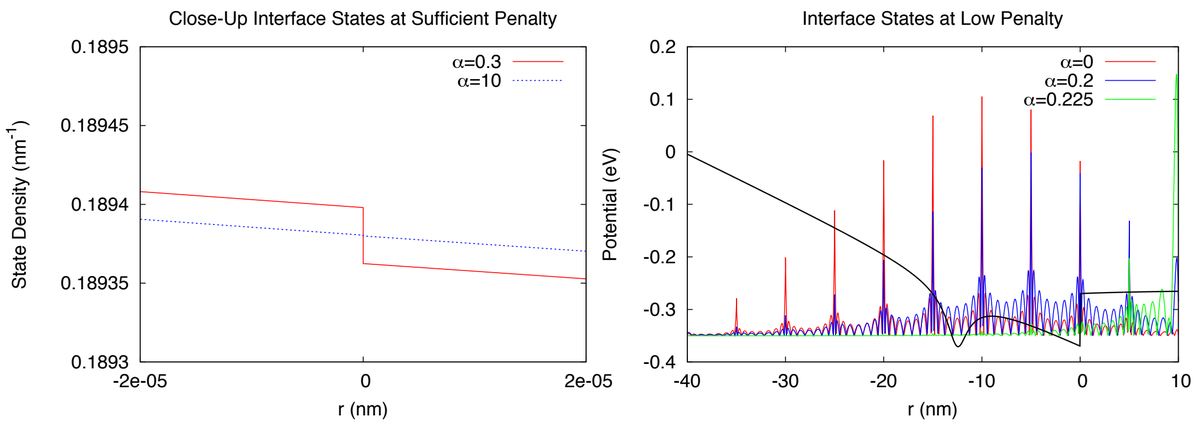
$$V^{HF}_\mu(\mathbf{r}) F_{i,\mu}(\mathbf{r}) = \sum_j \int d\mathbf{r}' \frac{|F_{j,\mu}(\mathbf{r}')|^2}{4\pi\epsilon_{Si}|\mathbf{r}-\mathbf{r}'|} - \sum_{j \neq i} \int d\mathbf{r}' \frac{F_{j,\mu}(\mathbf{r}') F_{i,\mu}(\mathbf{r}')}{4\pi\epsilon_{Si}|\mathbf{r}-\mathbf{r}'|}$$

Resolution-of-identity + IP Poisson solve facilitates perturbation theory

Results



(Left) line plot of solutions with 0.01% errors in energy
(Right) stabilized high order solutions for envelope functions



(Left) numerical discontinuity rigorously controllable
(Right) spurious solutions exist for small penalties

References

[1] Gamble, Jacobson, et al., Phys. Rev. B **91** (2015)
[2] Gamble, et al., App. Phys. Lett., **109**, (2016)
[3] Harvey-Collard, et al., arXiv:1512.01606
[4] Jock, et al., arXiv:1707.04357

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