

Title + Abstract

Useful Quantum Simulation: How classical and quantum computers can team up to tackle matters of substance

Quantum simulation is one of the most promising applications of quantum information technologies, aiming to elucidate the properties of interacting many-body systems spanning length scales from high energy, to nuclear, to chemical physics. Recent simulations of small systems on NISQ hardware and resource estimates for simulating classically intractable systems on general purpose hardware draw a line from where we are today to a world in which quantum hardware is seemingly the obvious choice for physical simulation of quantum systems. However, ongoing improvements to classical heuristics for simulating quantum systems mean that these goalposts will continue to move and might not have a sharply defined position in the first place. In this talk, I will argue that continued progress towards realizing useful quantum simulation capabilities should rely heavily on the exchange between these classical heuristics and quantum algorithms. I will discuss this in the context of my own work using classical high performance computing resources to simulate systems germane to fusion, catalysis, 2D materials, and even qubits.

Useful quantum simulation

How classical and quantum computers can team up
to tackle matters of substance

Andrew D. Baczewski

Quantum Computer Science
Center for Computing Research
Sandia National Laboratories



CQuIC Seminar Series,
University of New Mexico
November 8, 2018



Models and physics

$$\begin{aligned}
 & \text{1} \quad -\frac{1}{2}\partial_\nu g_\mu^a \partial_\nu g_\mu^a - g_s f^{abc} \partial_\mu g_\nu^a g_\mu^b g_\nu^c - \frac{1}{4}g_s^2 f^{abc} f^{ade} g_\mu^b g_\nu^c g_\mu^d g_\nu^e + \\
 & \quad \frac{1}{2}ig_s^2(\bar{q}_i^\sigma \gamma^\mu q_j^\sigma)g_\mu^a + \bar{G}^a \partial^2 G^a + g_s f^{abc} \partial_\mu \bar{G}^a G^b g_\mu^c - \partial_\nu W_\mu^+ \partial_\nu W_\mu^- - \\
 & \text{2} \quad M^2 W_\mu^+ W_\mu^- - \frac{1}{2}\partial_\nu Z_\mu^0 \partial_\nu Z_\mu^0 - \frac{1}{2c_w^2} M^2 Z_\mu^0 Z_\mu^0 - \frac{1}{2}\partial_\mu A_\nu \partial_\mu A_\nu - \frac{1}{2}\partial_\mu H \partial_\mu H - \\
 & \quad \frac{1}{2}m_h^2 H^2 - \partial_\mu \phi^+ \partial_\mu \phi^- - M^2 \phi^+ \phi^- - \frac{1}{2}\partial_\mu \phi^0 \partial_\mu \phi^0 - \frac{1}{2c_w^2} M \phi^0 \phi^0 - \beta_h \left[\frac{2M^2}{g^2} + \right. \\
 & \quad \left. \frac{2M}{g} H + \frac{1}{2}(H^2 + \phi^0 \phi^0 + 2\phi^+ \phi^-) \right] + \frac{2M^4}{g^2} \alpha_h - igc_w [\partial_\nu Z_\mu^0 (W_\mu^+ W_\nu^- - \\
 & \quad W_\nu^+ W_\mu^-) - Z_\nu^0 (W_\mu^+ \partial_\nu W_\mu^- - W_\mu^- \partial_\nu W_\mu^+) + Z_\mu^0 (W_\nu^+ \partial_\nu W_\mu^- - \\
 & \quad W_\nu^- \partial_\nu W_\mu^+)] - ig s_w [\partial_\nu A_\mu (W_\mu^+ W_\nu^- - W_\nu^+ W_\mu^-) - A_\nu (W_\mu^+ \partial_\nu W_\mu^- - \\
 & \quad W_\mu^- \partial_\nu W_\mu^+) + A_\mu (W_\nu^+ \partial_\nu W_\mu^- - W_\nu^- \partial_\nu W_\mu^+)] - \frac{1}{2}g^2 W_\mu^+ W_\mu^- W_\nu^+ W_\nu^- + \\
 & \quad \frac{1}{2}g^2 W_\mu^+ W_\nu^+ W_\mu^- W_\nu^- + g^2 c_w^2 (Z_\mu^0 W_\mu^+ Z_\nu^0 W_\nu^- - Z_\mu^0 Z_\nu^0 W_\mu^+ W_\nu^-) + \\
 & \quad g^2 s_w^2 (A_\mu W_\mu^+ A_\nu W_\nu^- - A_\mu A_\nu W_\mu^+ W_\nu^-) + g^2 s_w c_w [A_\mu Z_\nu^0 (W_\mu^+ W_\nu^- - \\
 & \quad W_\nu^+ W_\mu^-) - 2A_\mu Z_\mu^0 W_\nu^+ W_\nu^-] - g\alpha [H^3 + H\phi^0 \phi^0 + 2H\phi^+ \phi^-] - \\
 & \quad \frac{1}{8}g^2 \alpha_h [H^4 + (\phi^0)^4 + 4(\phi^+ \phi^-)^2 + 4(\phi^0)^2 \phi^+ \phi^- + 4H^2 \phi^+ \phi^- + 2(\phi^0)^2 H^2] - \\
 & \quad g M W_\mu^+ W_\mu^- H - \frac{1}{2}g \frac{M}{c_w^2} Z_\mu^0 Z_\mu^0 H - \frac{1}{2}ig [W_\mu^+ (\phi^0 \partial_\mu \phi^- - \phi^- \partial_\mu \phi^0) - \\
 & \quad W_\mu^- (\phi^0 \partial_\mu \phi^+ - \phi^+ \partial_\mu \phi^0)] + \frac{1}{2}g [W_\mu^+ (H \partial_\mu \phi^- - \phi^- \partial_\mu H) - W_\mu^- (H \partial_\mu \phi^+ - \\
 & \quad \phi^+ \partial_\mu H)] + \frac{1}{2}g \frac{1}{c_w} (Z_\mu^0 (H \partial_\mu \phi^0 - \phi^0 \partial_\mu H) - ig \frac{s_w^2}{c_w} M Z_\mu^0 (W_\mu^+ \phi^- - W_\mu^- \phi^+) + \\
 & \quad ig s_w M A_\mu (W_\mu^+ \phi^- - W_\mu^- \phi^+) - ig \frac{1-2c_w^2}{2c_w} Z_\mu^0 (\phi^+ \partial_\mu \phi^- - \phi^- \partial_\mu \phi^+) + \\
 & \quad ig s_w A_\mu (\phi^+ \partial_\mu \phi^- - \phi^- \partial_\mu \phi^+) - \frac{1}{4}g^2 W_\mu^+ W_\mu^- [H^2 + (\phi^0)^2 + 2\phi^+ \phi^-] - \\
 & \quad \frac{1}{4}g^2 \frac{1}{c_w^2} Z_\mu^0 Z_\mu^0 [H^2 + (\phi^0)^2 + 2(2s_w^2 - 1)\phi^+ \phi^-] - \frac{1}{2}g^2 \frac{s_w^2}{c_w} Z_\mu^0 \phi^0 (W_\mu^+ \phi^- + \\
 & \quad W_\mu^- \phi^+) - \frac{1}{2}ig^2 \frac{s_w^2}{c_w} Z_\mu^0 H (W_\mu^+ \phi^- - W_\mu^- \phi^+) + \frac{1}{2}g^2 s_w A_\mu \phi^0 (W_\mu^+ \phi^- + \\
 & \quad W_\mu^- \phi^+) + \frac{1}{2}ig^2 s_w A_\mu H (W_\mu^+ \phi^- - W_\mu^- \phi^+) - g^2 \frac{s_w}{c_w} (2c_w^2 - 1) Z_\mu^0 A_\mu \phi^+ \phi^- - \\
 & \quad g^4 s_w^2 A_\mu A_\mu \phi^+ \phi^- - \bar{e}^\lambda (\gamma \partial + m_e^\lambda) e^\lambda - \bar{\nu}^\lambda \gamma \partial \nu^\lambda - \bar{u}_j^\lambda (\gamma \partial + m_u^\lambda) u_j^\lambda - \\
 & \text{3} \quad \bar{d}_j^\lambda (\gamma \partial + m_d^\lambda) d_j^\lambda + ig s_w A_\mu [-(\bar{e}^\lambda \gamma^\mu e^\lambda) + \frac{2}{3}(\bar{u}_j^\lambda \gamma^\mu u_j^\lambda) - \frac{1}{3}(\bar{d}_j^\lambda \gamma^\mu d_j^\lambda)] + \\
 & \quad \frac{ig}{4c_w} Z_\mu^0 [(\bar{\nu}^\lambda \gamma^\mu (1 + \gamma^5) \nu^\lambda) + (e^\lambda \gamma^\mu (4s_w^2 - 1 - \gamma^5) e^\lambda) + (\bar{u}_j^\lambda \gamma^\mu (\frac{4}{3}s_w^2 - \\
 & \quad 1 - \gamma^5) u_j^\lambda) + (\bar{d}_j^\lambda \gamma^\mu (1 - \frac{8}{3}s_w^2 - \gamma^5) d_j^\lambda)] + \frac{ig}{2\sqrt{2}} W_\mu^+ [(\bar{\nu}^\lambda \gamma^\mu (1 + \gamma^5) e^\lambda) + \\
 & \quad (\bar{u}_j^\lambda \gamma^\mu (1 + \gamma^5) C_{\lambda\kappa} d_j^\kappa)] + \frac{ig}{2\sqrt{2}} W_\mu^- [(\bar{e}^\lambda \gamma^\mu (1 + \gamma^5) \nu^\lambda) + (\bar{d}_j^\kappa C_{\lambda\kappa}^\dagger \gamma^\mu (1 + \\
 & \quad \gamma^5) u_j^\lambda)] + \frac{ig}{2\sqrt{2}} \frac{m_\lambda^2}{M} [-\phi^+ (\bar{\nu}^\lambda (1 - \gamma^5) e^\lambda) + \phi^- (\bar{e}^\lambda (1 + \gamma^5) \nu^\lambda)] - \\
 & \text{4} \quad \frac{g}{2} \frac{m_\lambda^2}{M} [H(\bar{e}^\lambda e^\lambda) + i\phi^0 (\bar{e}^\lambda \gamma^5 e^\lambda)] + \frac{ig}{2M\sqrt{2}} \phi^+ [-m_d^\lambda (\bar{u}_j^\lambda C_{\lambda\kappa} (1 - \gamma^5) d_j^\kappa) + \\
 & \quad m_u^\lambda (\bar{u}_j^\lambda C_{\lambda\kappa} (1 + \gamma^5) d_j^\kappa) + \frac{ig}{2M\sqrt{2}} \phi^- [m_d^\lambda (\bar{d}_j^\lambda C_{\lambda\kappa}^\dagger (1 + \gamma^5) u_j^\kappa) - m_u^\lambda (\bar{d}_j^\lambda C_{\lambda\kappa}^\dagger (1 - \\
 & \quad \gamma^5) u_j^\kappa) - \frac{g}{2} \frac{m_\lambda^2}{M} H(\bar{u}_j^\lambda u_j^\lambda) - \frac{g}{2} \frac{m_\lambda^2}{M} H(\bar{d}_j^\lambda d_j^\lambda) + \frac{ig}{2} \frac{m_\lambda^2}{M} \phi^0 (\bar{u}_j^\lambda \gamma^5 u_j^\lambda) - \\
 & \quad \frac{ig}{2} \frac{m_\lambda^2}{M} \phi^0 (\bar{d}_j^\lambda \gamma^5 d_j^\lambda)] + \bar{X}^+ (\partial^2 - M^2) X^+ + \bar{X}^- (\partial^2 - M^2) X^- + \bar{X}^0 (\partial^2 - \\
 & \text{5} \quad \frac{M^2}{c_w^2}) X^0 + \bar{Y} \partial^2 Y + igc_w W_\mu^+ (\partial_\mu \bar{X}^0 X^- - \partial_\mu \bar{X}^+ X^0) + ig s_w W_\mu^+ (\partial_\mu \bar{Y} X^- - \\
 & \quad \partial_\mu \bar{X}^+ Y) + igc_w W_\mu^- (\partial_\mu \bar{X}^- X^0 - \partial_\mu \bar{X}^0 X^+) + ig s_w W_\mu^- (\partial_\mu \bar{X}^- Y - \\
 & \quad \partial_\mu \bar{Y} X^+) + igc_w Z_\mu^0 (\partial_\mu \bar{X}^+ X^+ - \partial_\mu \bar{X}^- X^-) + ig s_w A_\mu (\partial_\mu \bar{X}^+ X^+ - \\
 & \quad \partial_\mu \bar{X}^- X^-) - \frac{1}{2}gM [\bar{X}^+ X^+ H + \bar{X}^- X^- H + \frac{1}{c_w^2} \bar{X}^0 X^0 H] + \\
 & \quad \frac{1-2c_w^2}{2c_w} igM [\bar{X}^+ X^0 \phi^+ - \bar{X}^- X^0 \phi^-] + \frac{1}{2c_w} igM [\bar{X}^0 X^- \phi^+ - \bar{X}^+ X^+ \phi^-] + \\
 & \quad igMs_w [\bar{X}^0 X^- \phi^+ - \bar{X}^+ X^+ \phi^-] + \frac{1}{2}igM [\bar{X}^+ X^+ \phi^0 - \bar{X}^- X^- \phi^0]
 \end{aligned}$$

Standard model Lagrangian

Integrating out quarks and radioactive decay,
and focusing only on the interaction of **nuclei** and **electrons**

$$\sum_{i=1}^{N_e} \left(-\frac{\nabla_i^2}{2} + V_{ext}(\mathbf{r}_i) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} \right) + \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Quantum chemical Hamiltonian

Integrating out tightly bound core electrons,
and focusing only on the mutual interaction of **strongly correlated electrons**

$$t \sum_{\langle i,j \rangle, \sigma} \left(\hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} + \text{h.c.} \right) + U \sum_i \hat{c}_{i,\uparrow}^\dagger \hat{c}_{i,\uparrow} \hat{c}_{i,\downarrow}^\dagger \hat{c}_{i,\downarrow}$$

Single orbital Fermi-Hubbard model

Just because we can efficiently specify a model,
does not mean that we can efficiently solve it

This isn't a surprise

Quantum Mechanics of Many-Electron Systems.

By P. A. M. DIRAC, St. John's College, Cambridge.

(Communicated by R. H. Fowler, F.R.S.—Received March 12, 1929.)

§ 1. *Introduction.*

The general theory of quantum mechanics is now almost complete, the imperfections that still remain being in connection with the exact fitting in of the theory with relativity ideas. These give rise to difficulties only when high-speed particles are involved, and are therefore of no importance in the consideration of atomic and molecular structure and ordinary chemical reactions, in which it is, indeed, usually sufficiently accurate if one neglects relativity variation of mass with velocity and assumes only Coulomb forces between the various electrons and atomic nuclei. The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble. It therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.



Quantum simulation

1982

“...because nature isn’t classical, dammit, and if you want to make a simulation of nature, you’d better make it quantum mechanical, and by golly it’s a wonderful problem because it doesn’t look so easy.”

1996

Universal Quantum Simulators

Seth Lloyd

Feynman’s 1982 conjecture, that quantum computers can be programmed to simulate any local quantum system, is shown to be correct.

1997 & 1999

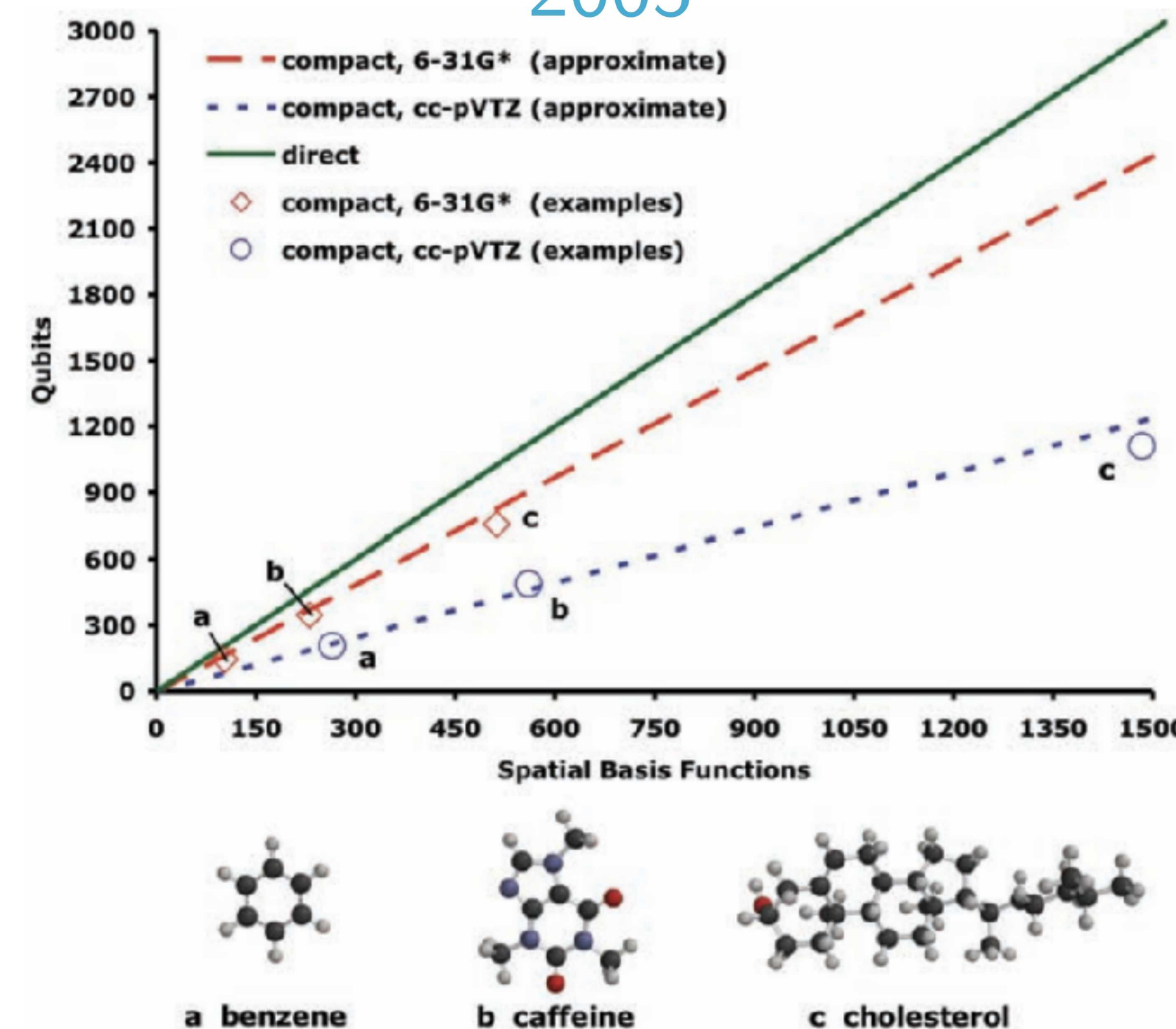
Quantum Algorithm Providing Exponential Speed Increase for Finding Eigenvalues and Eigenvectors

Daniel S. Abrams and Seth Lloyd
Phys. Rev. Lett. **83**, 5162 – Published 13 December 1999

Simulation of Many-Body Fermi Systems on a Universal Quantum Computer

Daniel S. Abrams and Seth Lloyd
Phys. Rev. Lett. **79**, 2586 – Published 29 September 1997

2005

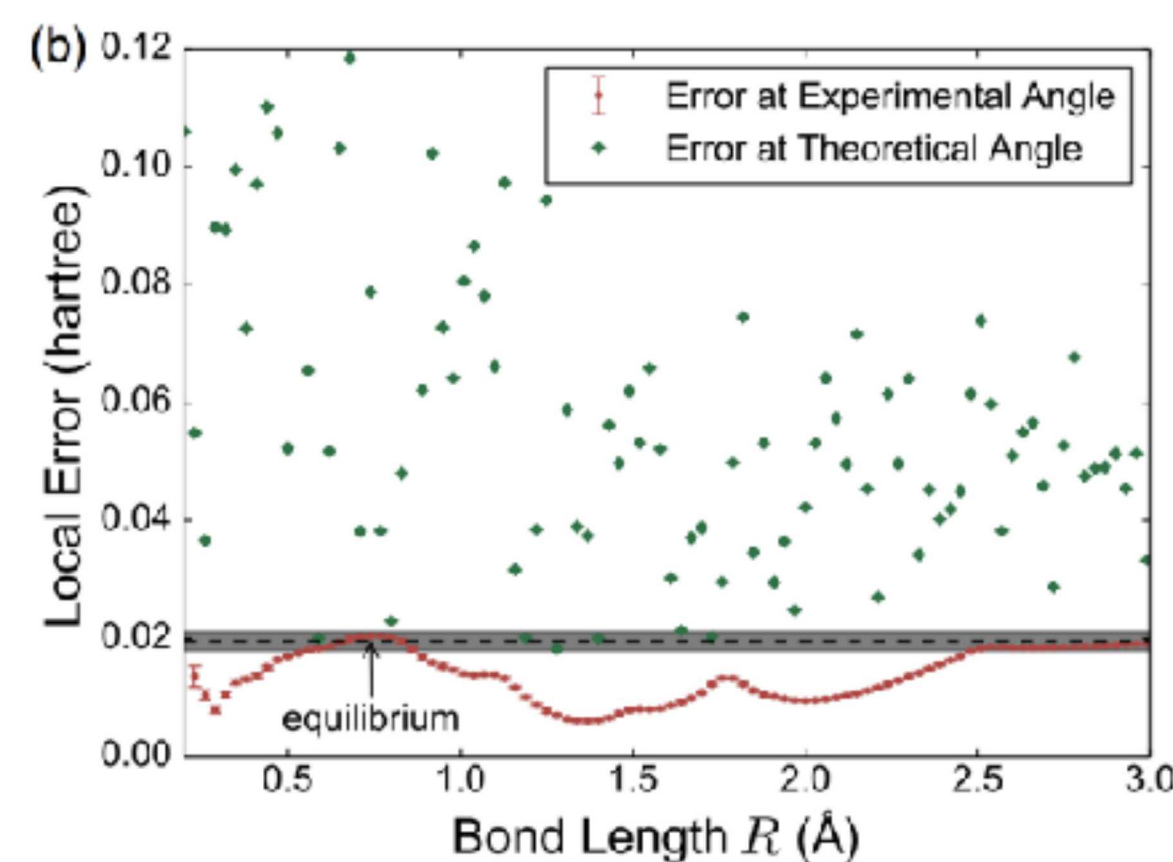
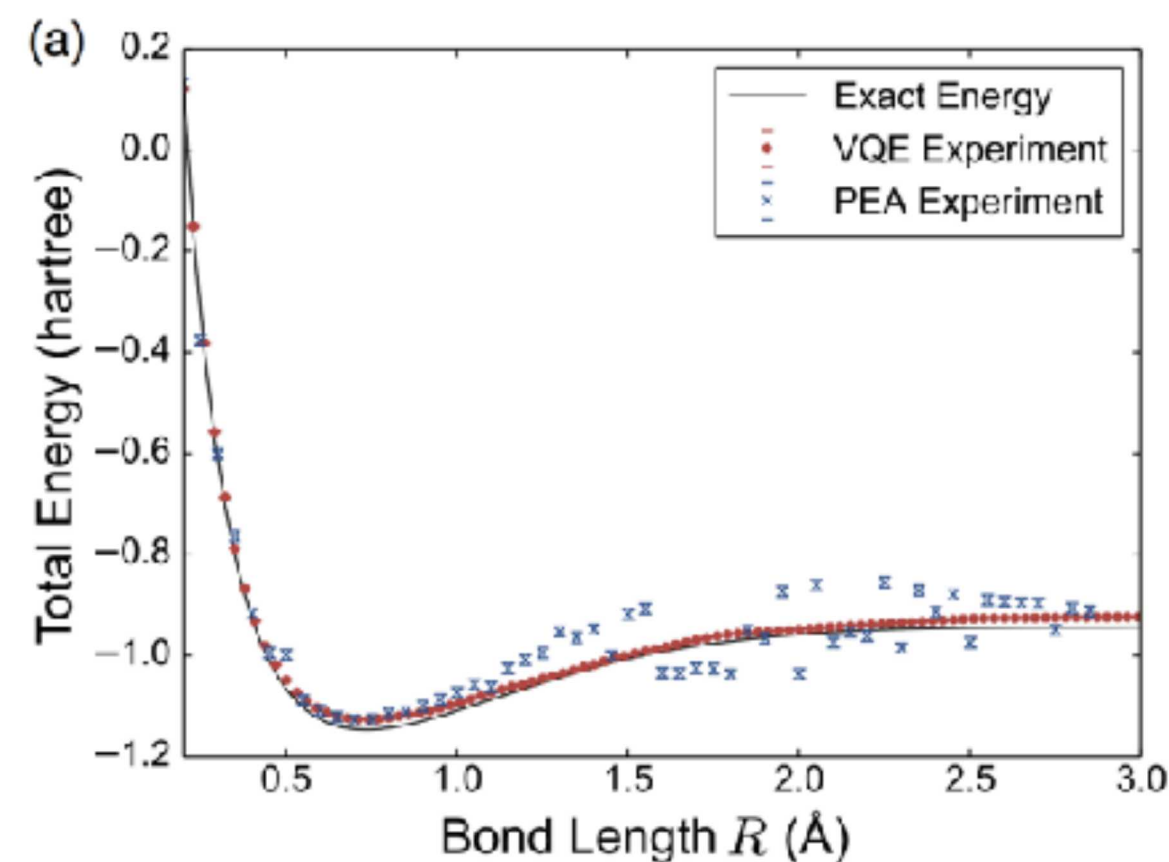
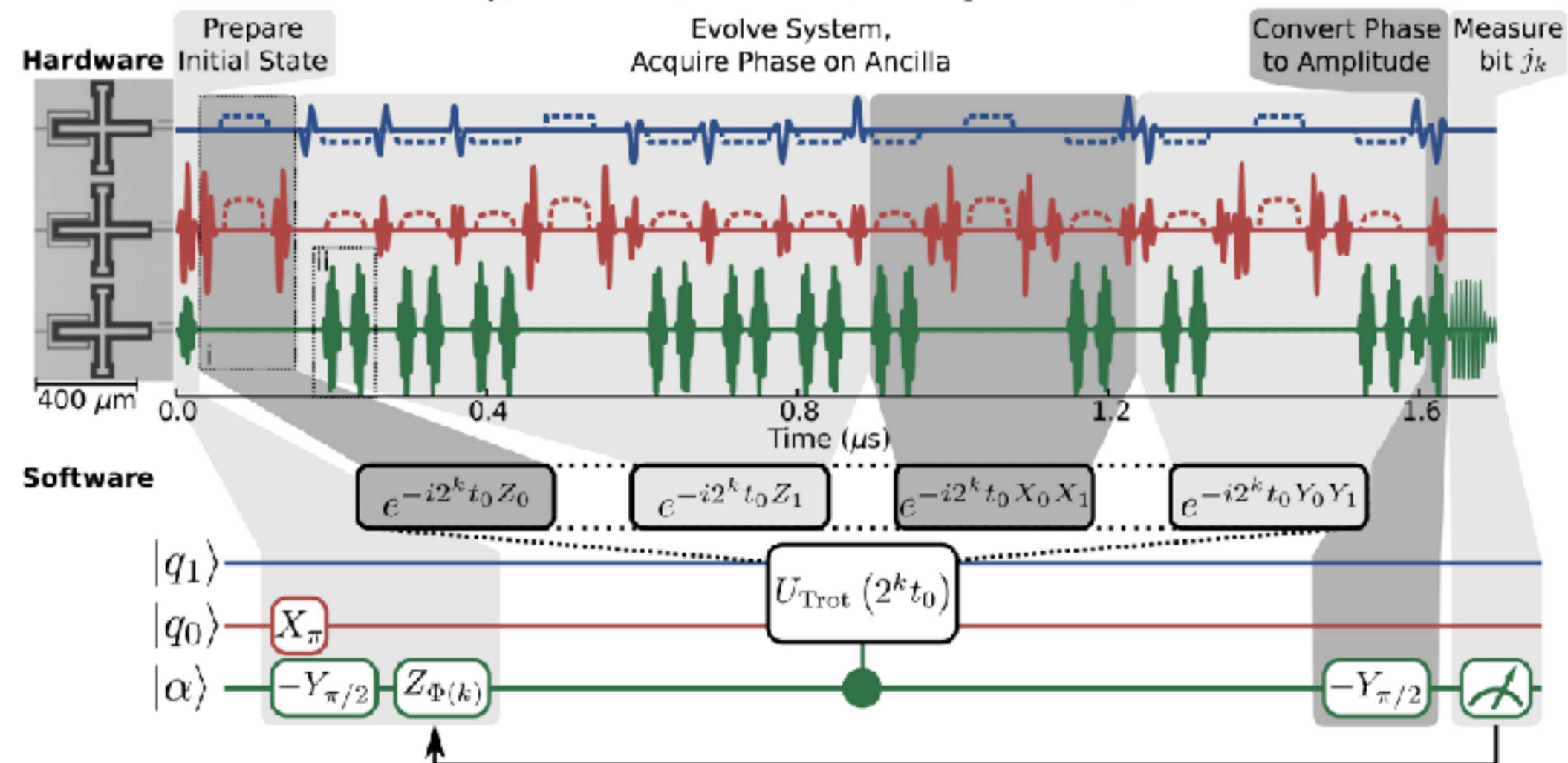


Simulations on NISQ hardware

PHYSICAL REVIEW X 6, 031007 (2016)

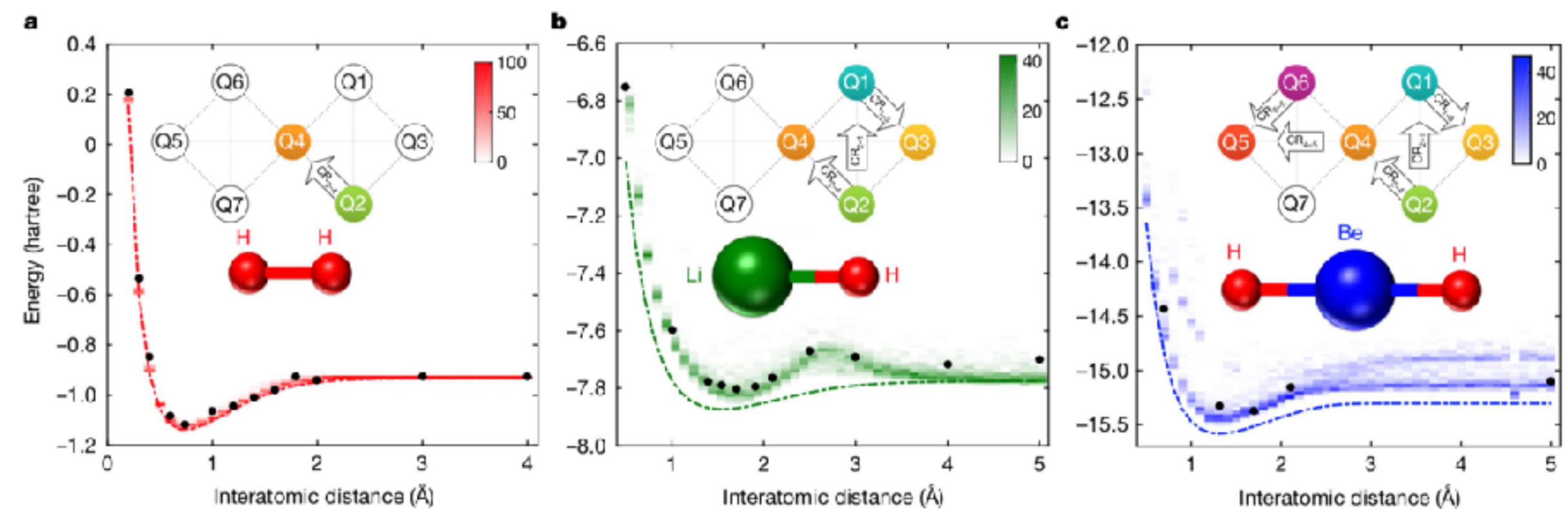
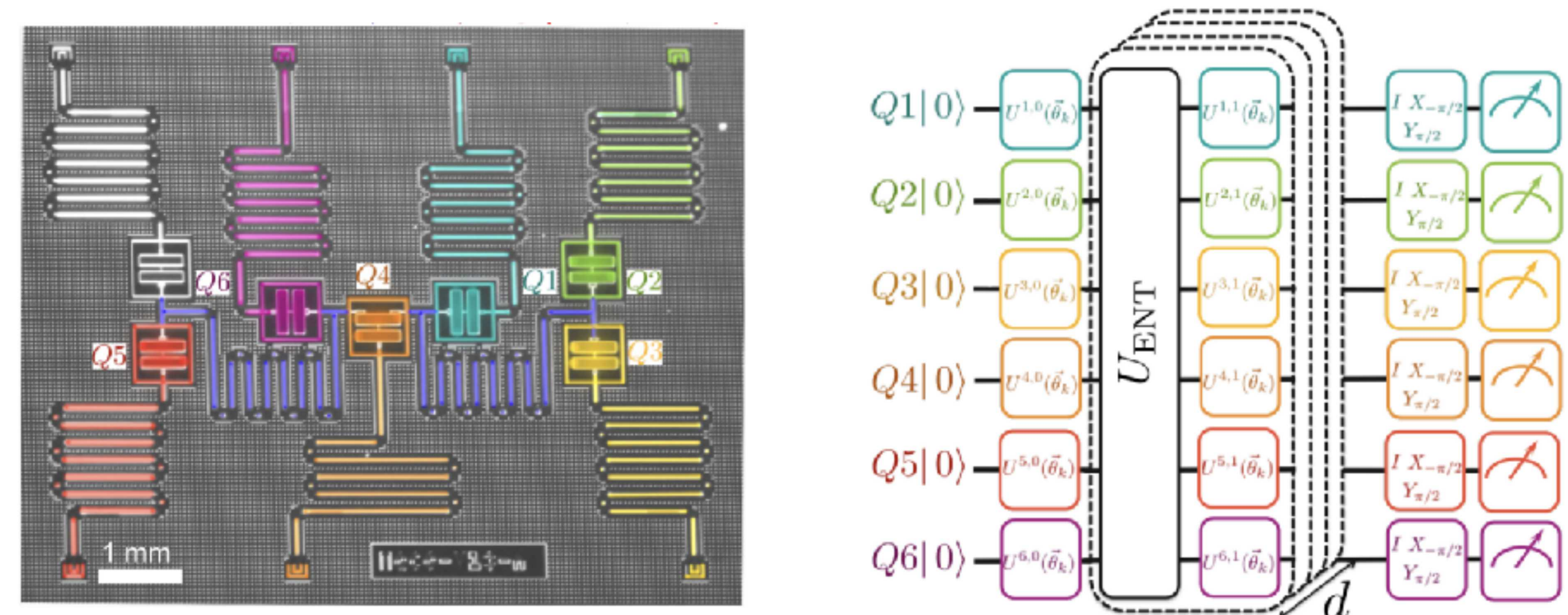
Scalable Quantum Simulation of Molecular Energies

P. J. J. O'Malley,^{1,*} R. Babbush,^{2,†} I. D. Kivlichan,³ J. Romero,³ J. R. McClean,⁴ R. Barends,⁵ J. Kelly,⁵ P. Roushan,⁵ A. Tranter,^{6,7} N. Ding,² B. Campbell,¹ Y. Chen,⁵ Z. Chen,¹ B. Chiaro,¹ A. Dunsworth,¹ A. G. Fowler,⁵ E. Jeffrey,⁵ E. Lucero,⁵ A. Megrant,⁵ J. Y. Mutus,⁵ M. Neeley,⁵ C. Neill,¹ C. Quintana,¹ D. Sank,⁵ A. Vainsencher,¹ J. Wenner,¹ T. C. White,⁵ P. V. Coveney,⁷ P. J. Love,⁶ H. Neven,² A. Aspuru-Guzik,³ and J. M. Martinis^{5,1,‡}



Hardware-efficient variational quantum eigensolver for small molecules and quantum magnets

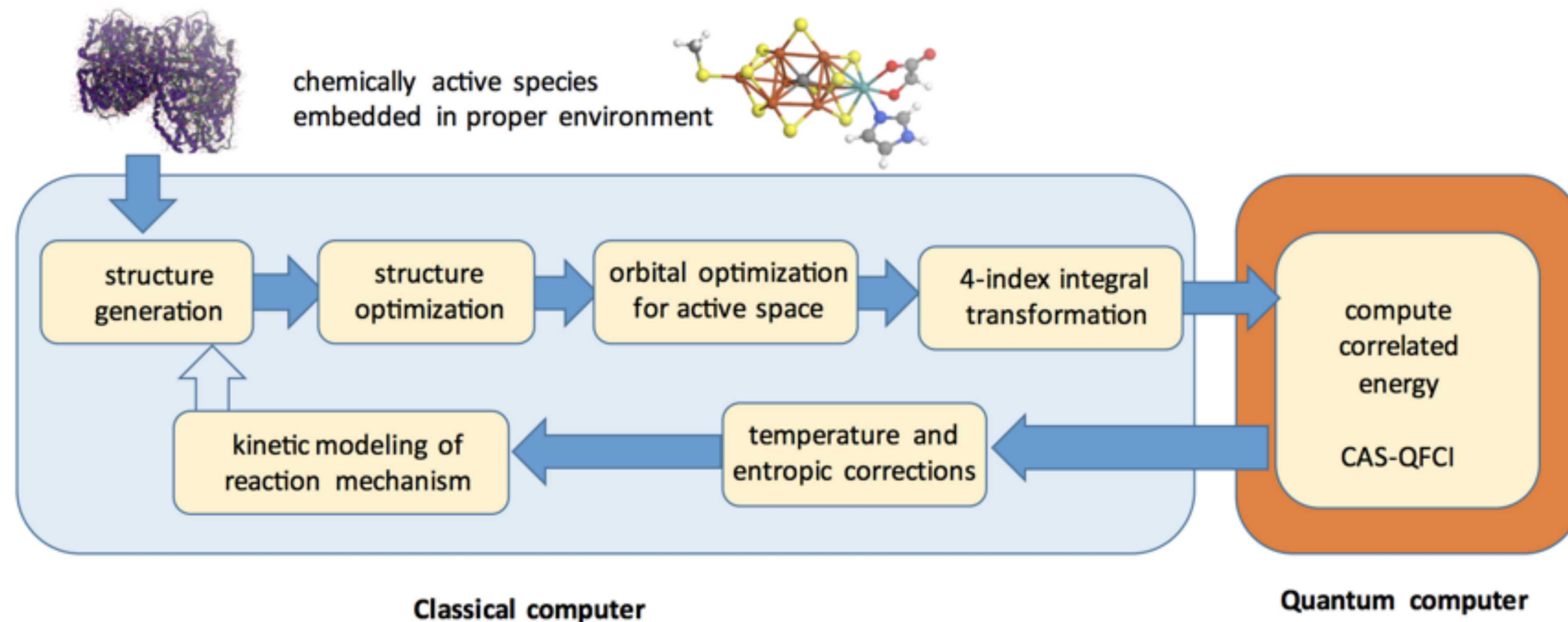
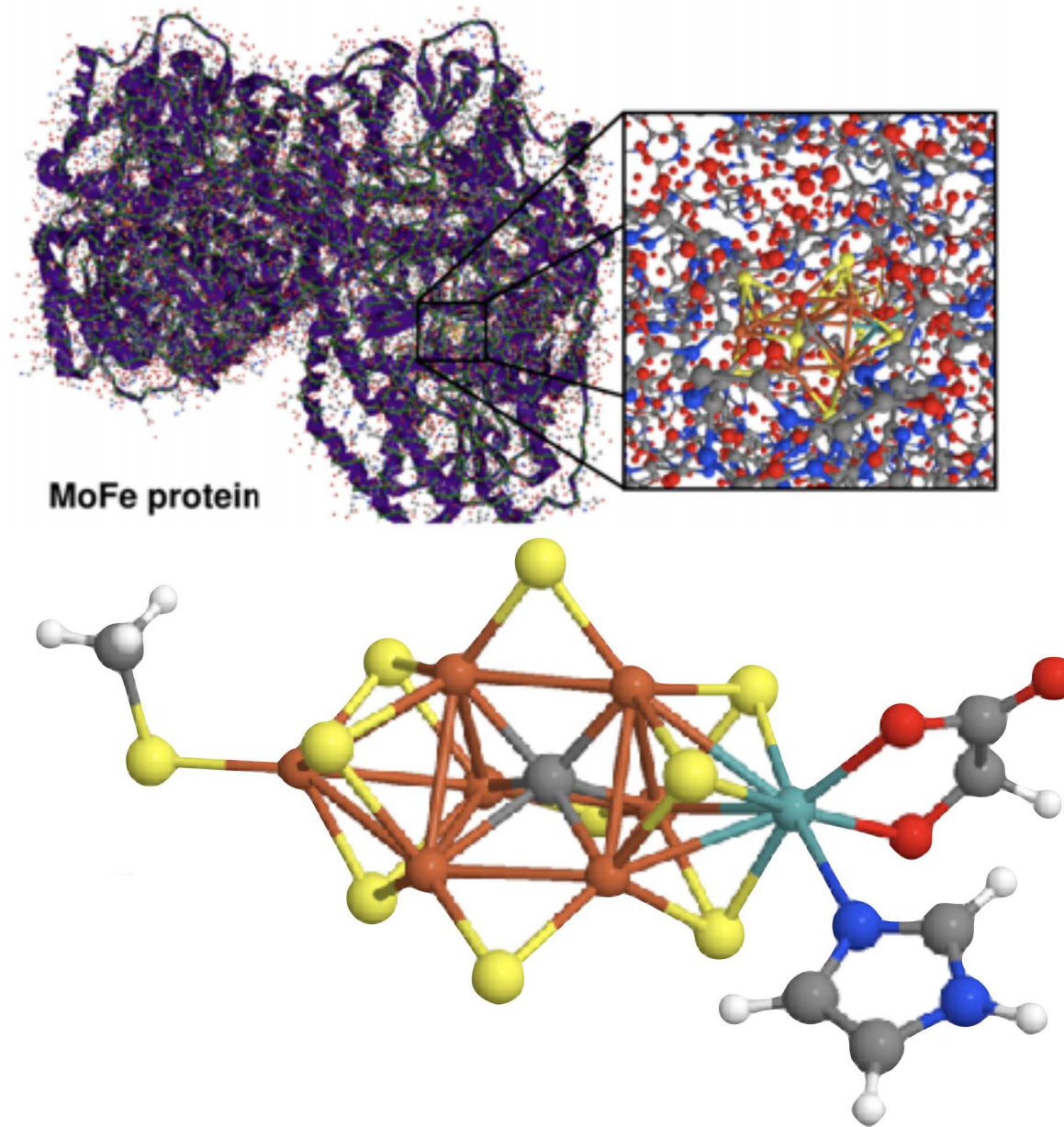
Abhinav Kandala^{1,*}, Antonio Mezzacapo^{1,*}, Kristan Temme¹, Maika Takita¹, Markus Brink¹, Jerry M. Chow¹ & Jay M. Gambetta¹



Simulations on general purpose hardware

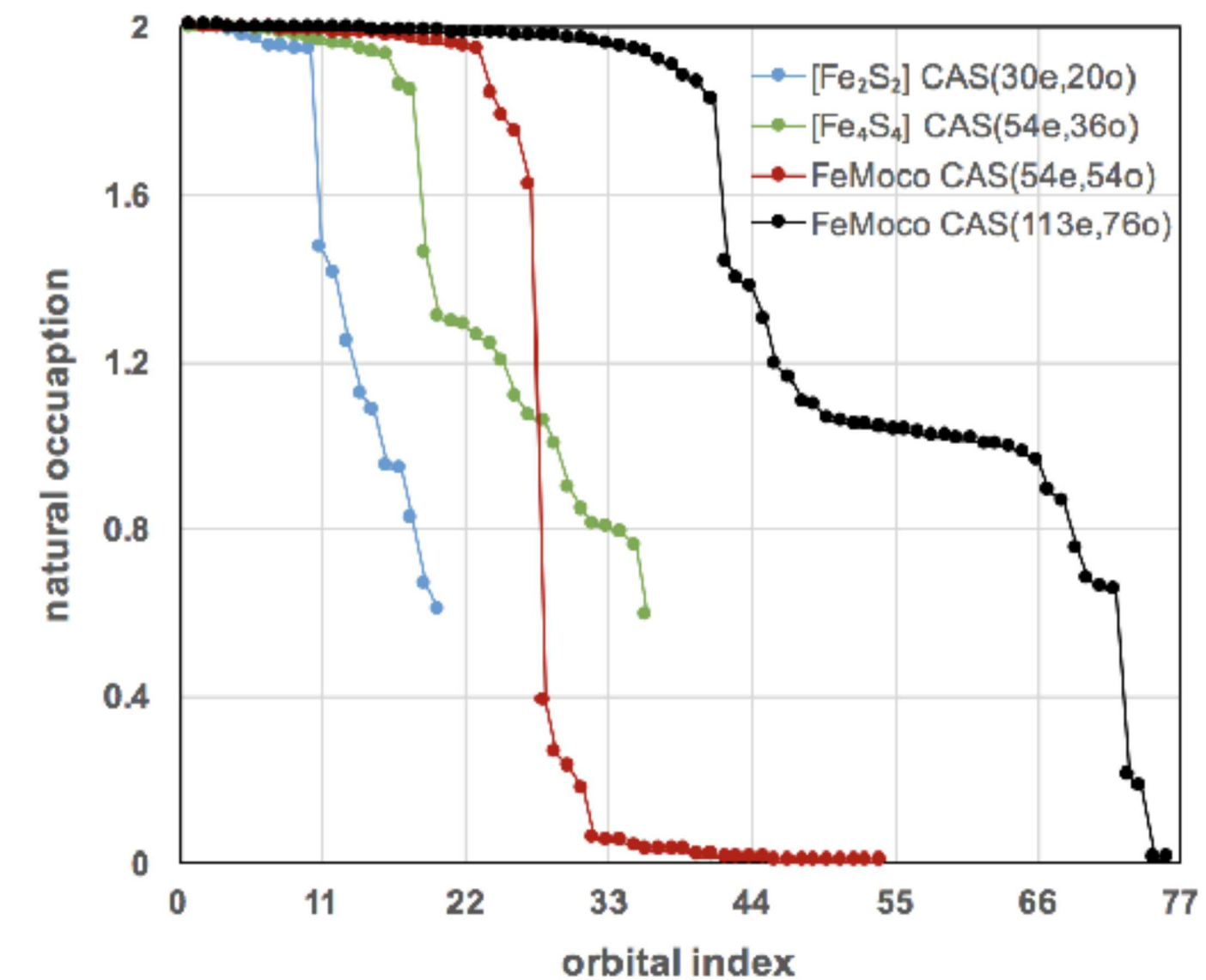
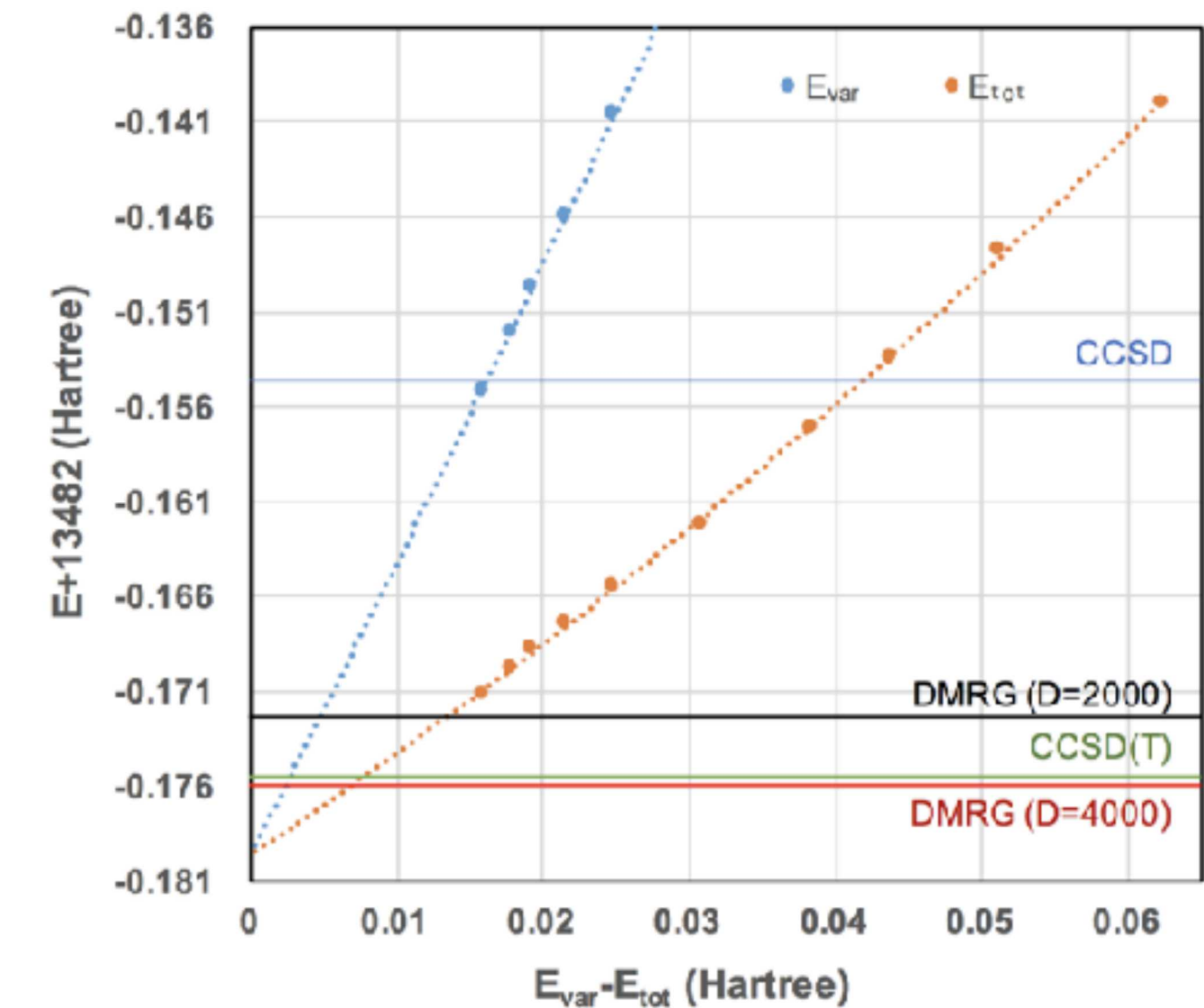
Elucidating reaction mechanisms on quantum computers

Markus Reiher^{a,1}, Nathan Wiebe^{b,1}, Krysta M. Svore^b, Dave Wecker^b, and Matthias Troyer^{b,c,2}



The electronic complexity of the ground-state of the FeMo cofactor of nitrogenase as relevant to quantum simulations

Zhendong Li,¹ Junhao Li,² Nikesh S. Dattani,³ C. J. Umrigar,² and Garnet Kin-Lic Chan¹



Keep reading

Quantum Mechanics of Many-Electron Systems.

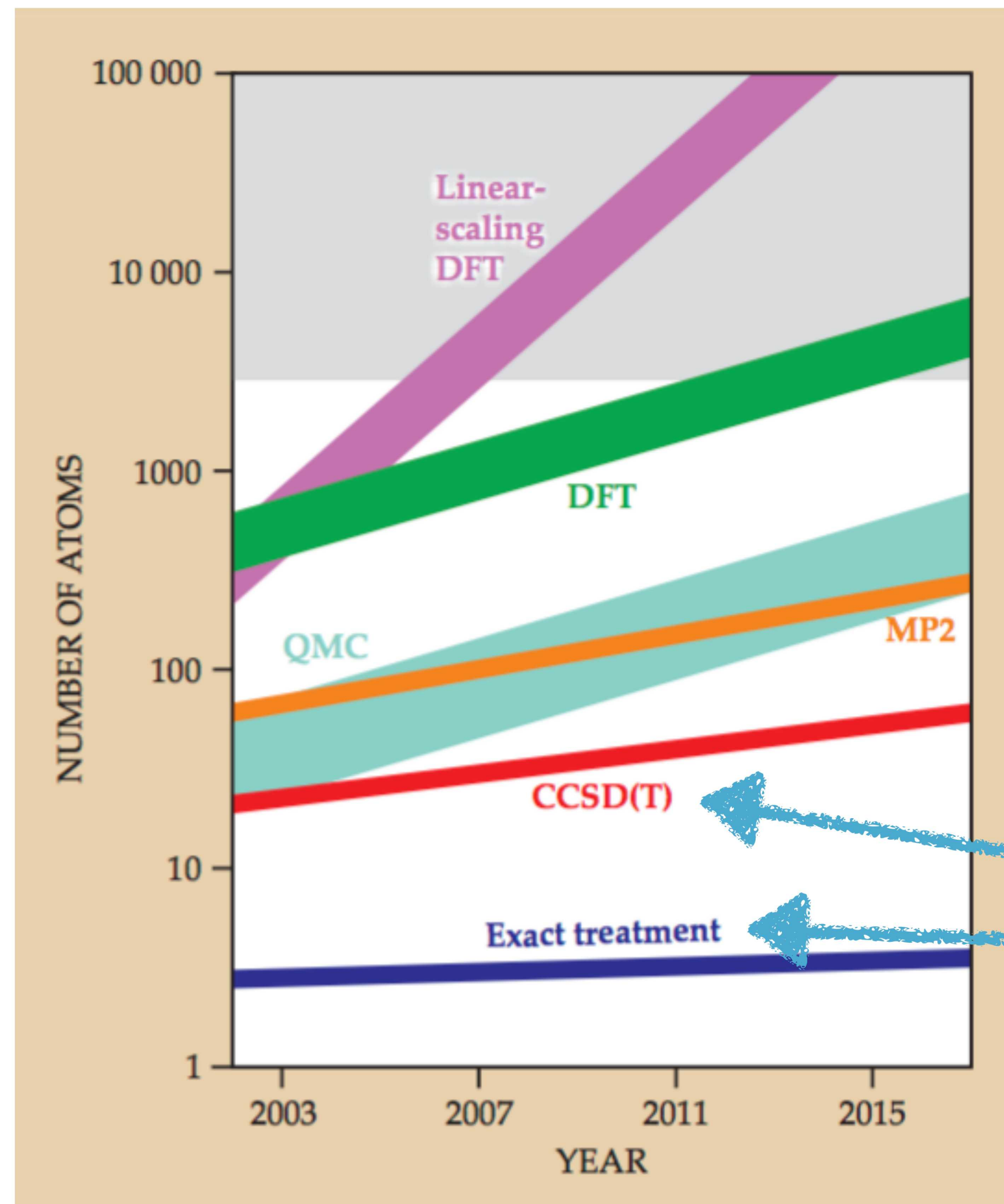
By P. A. M. DIRAC, St. John's College, Cambridge.

(Communicated by R. H. Fowler, F.R.S.—Received March 12, 1929.)

§ 1. *Introduction.*

The general theory of quantum mechanics is now almost complete, the imperfections that still remain being in connection with the exact fitting in of the theory with relativity ideas. These give rise to difficulties only when high-speed particles are involved, and are therefore of no importance in the consideration of atomic and molecular structure and ordinary chemical reactions, in which it is, indeed, usually sufficiently accurate if one neglects relativity variation of mass with velocity and assumes only Coulomb forces between the various electrons and atomic nuclei. The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble. It therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.

Classical heuristics



*Head-Gordon and Artacho
Physics Today (2008)*

* Classical heuristics portend how we will use quantum simulation, *this talk will describe some of those heuristics and how they interact with near-term quantum simulation research*

Virtuous cycle: classical heuristics can improve quantum hardware and quantum hardware can improve classical heuristics

Expensive classical heuristics are typically *less accurate*, and we typically *don't* know how accurate they are**

...but the actual “competition” is *90 years of classical heuristics*

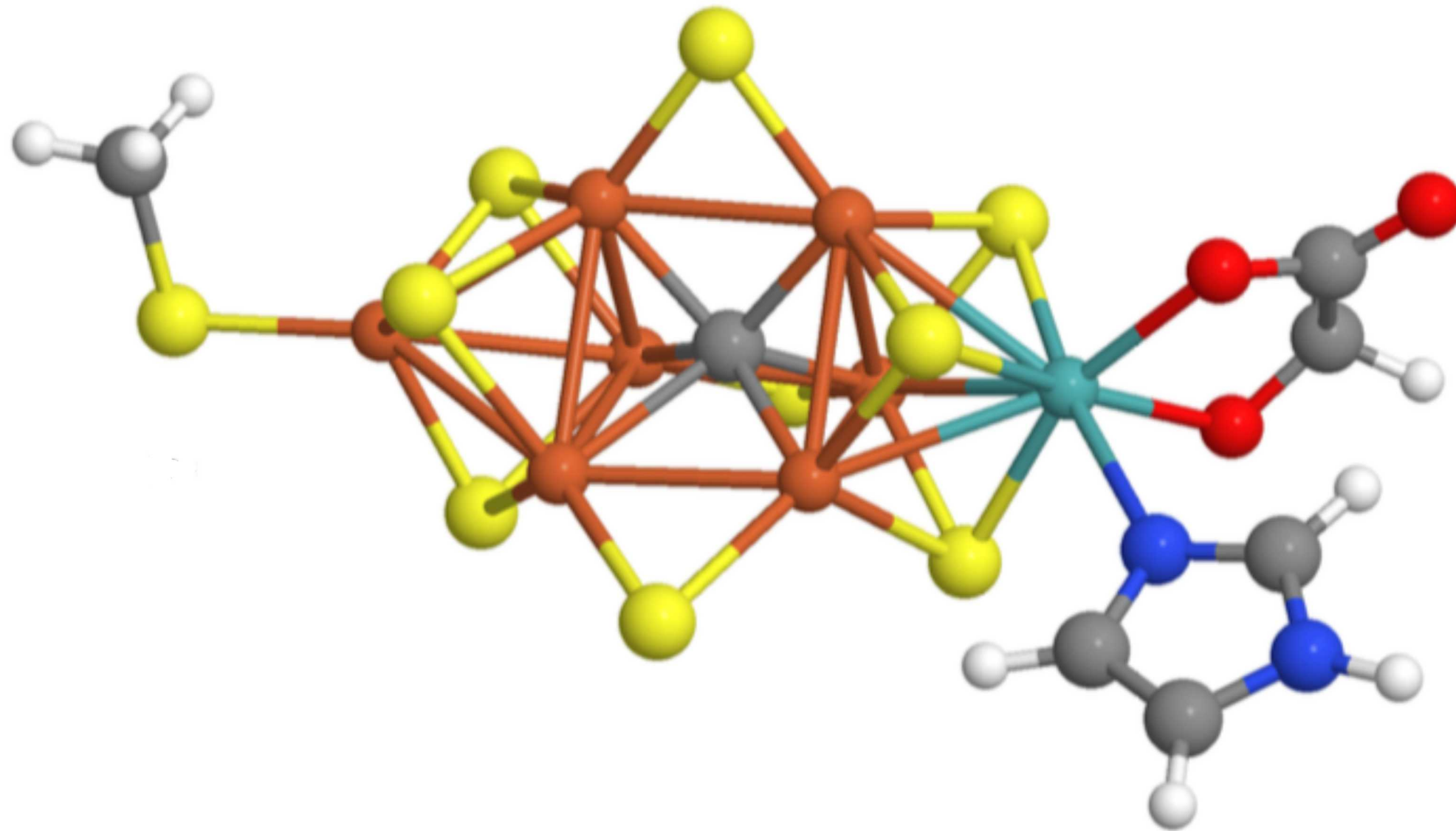
When you see a talk on quantum simulation, this is frequently the classical “competition”

* This article preceded the widespread use of tensor network methods in first principles modeling

** If we did it would have deep ramifications for computational complexity

Problem statement

$$H = \sum_{i=1}^{N_e} \left(-\frac{\nabla_i^2}{2} + V_{ext}(\mathbf{r}_i) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} \right) + \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$



Reiher, et al., PNAS (2017)

- “First principles” modeling, we know:
 - Positions of N_n semiclassical nuclei, $\mathbf{R}_a$
 - Charges of those nuclei, Z_a
 - Total number of electrons, N_e
- We want to know:
 - Ground state $H|\Psi_0\rangle = E_0|\Psi_0\rangle$
 - Excited states $H|\Psi_n\rangle = E_n|\Psi_n\rangle$
 - Observables $\langle\Psi_0|O|\Psi_0\rangle$
 - Response functions $G(E) = \lim_{\eta \rightarrow 0^+} (H - E + i\eta)^{-1}$

Density functional theory (DFT)

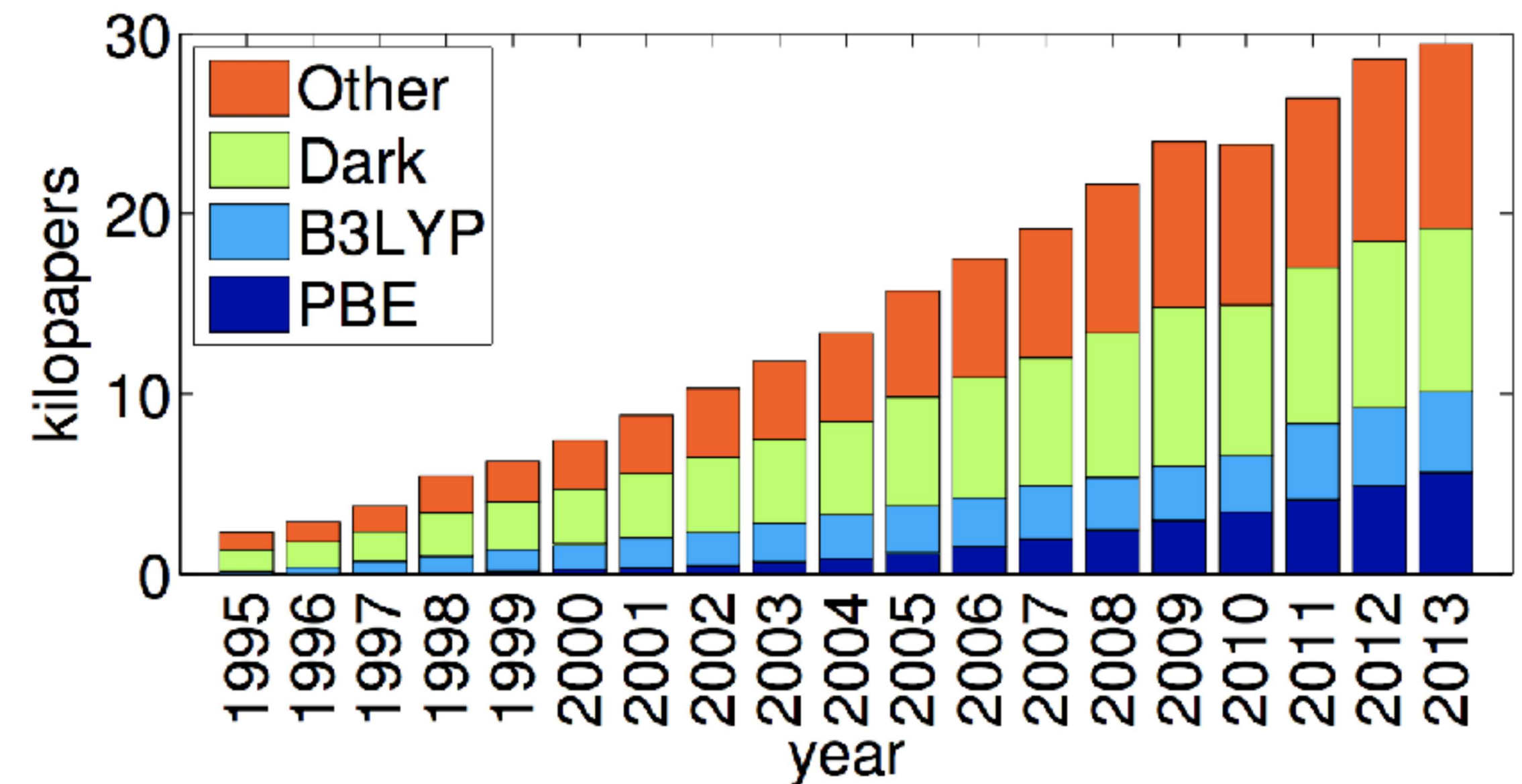
The most popular approach to materials simulation (and *large* chemical simulations)

Pros:

- Non-perturbative, requires exchange-correlation functional
- Cubic-scaling with system size, typically decides cost vs. accuracy dilemma
- Quadratic- and linear-scaling variants exist, but the cost prefactors are still quite large

Cons:

- Non-perturbative, requires exchange-correlation functional
- Finding that functional is QMA-Complete (*Schuch and Verstraete, Nat. Phys. (2009)*)
- There is no real wave function, the only meaningful observables are density functionals



*Pribram-Jones, Gross, Burke
Ann. Rev. Phys. Chem. (2015)*

DFT in a bit more detail

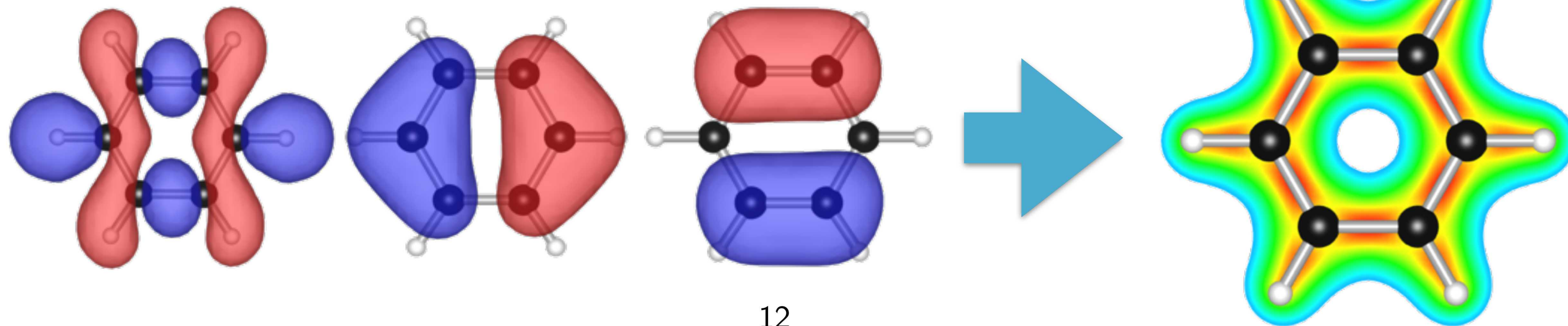
$$H = \sum_{i=1}^{N_e} \left(-\frac{\nabla_i^2}{2} + V_{ext}(\mathbf{r}_i) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} \right) + \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Find a **density** that minimizes a **density functional** for the **total energy**

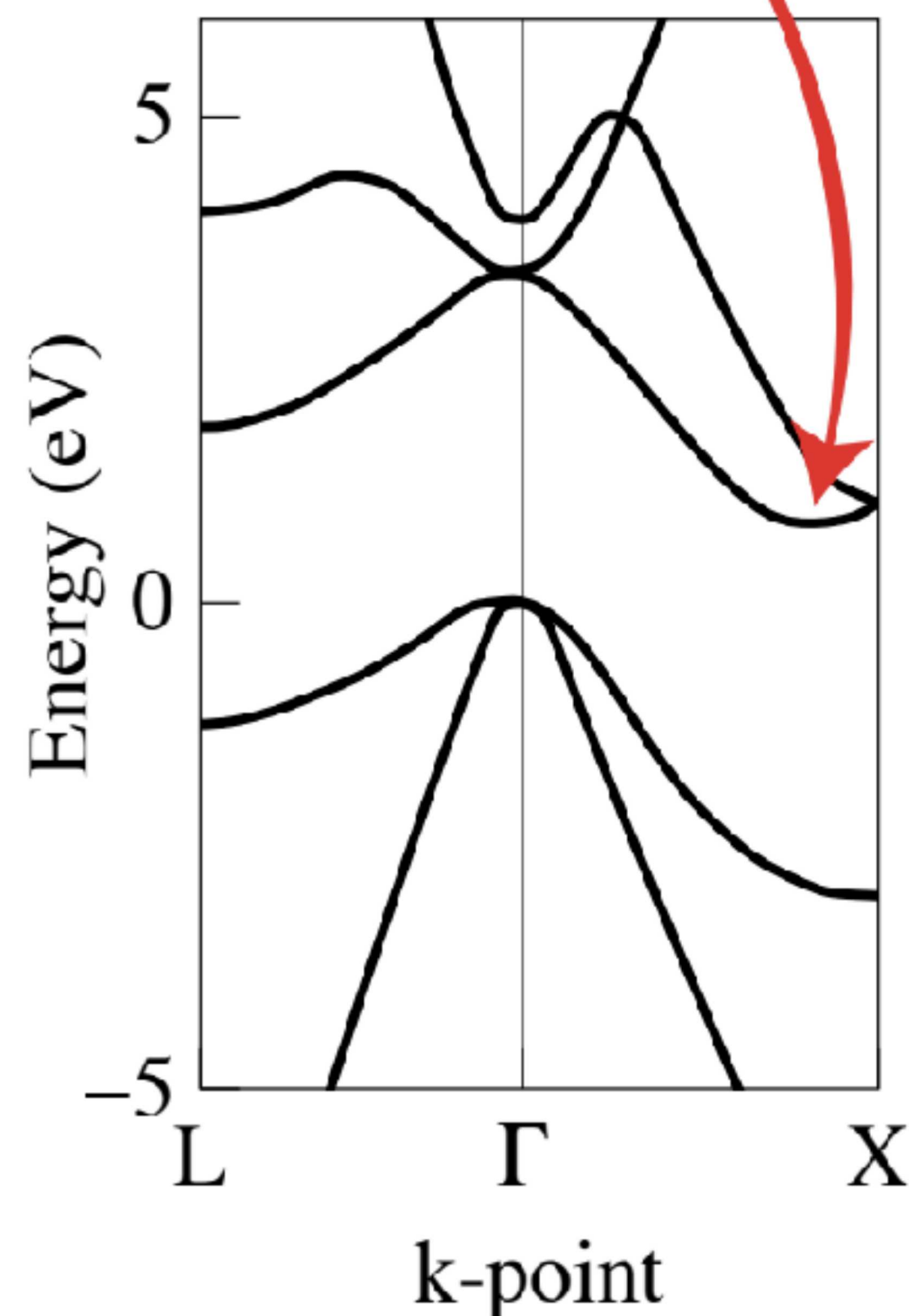
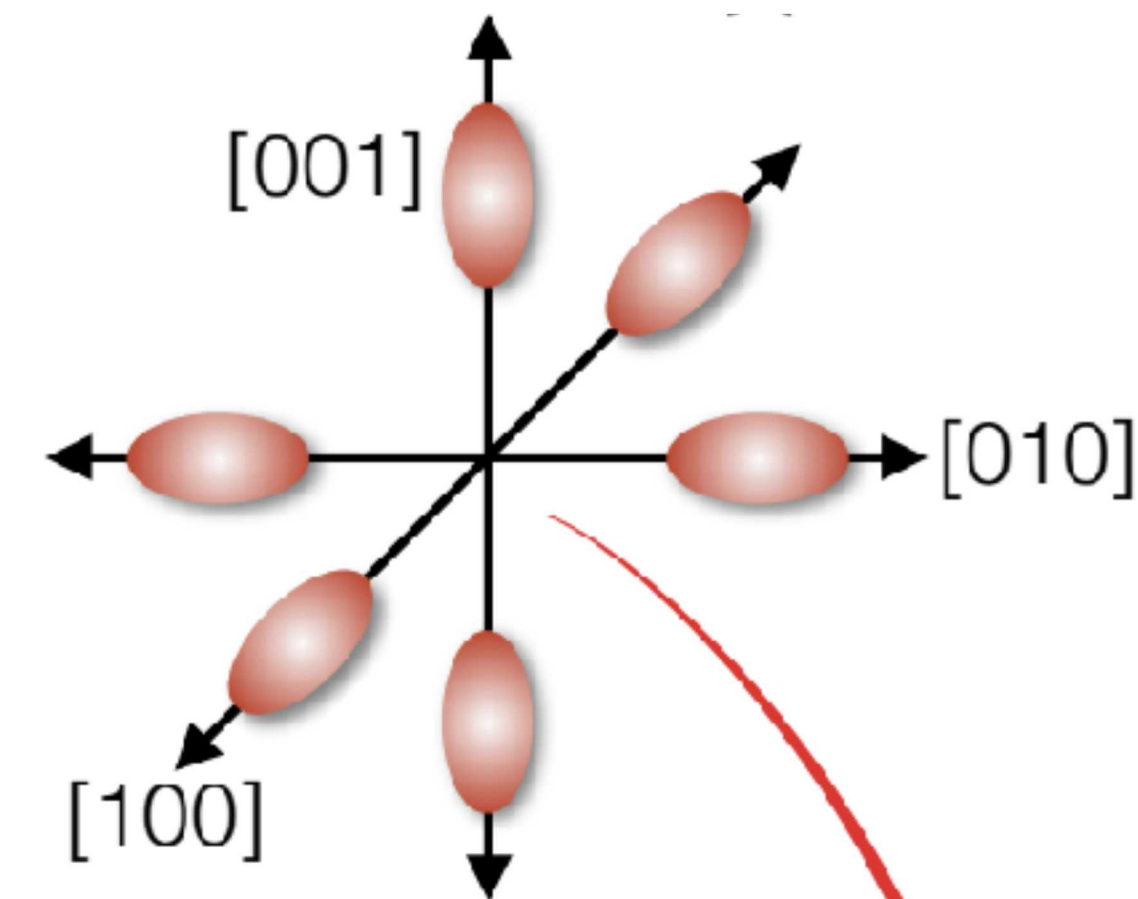
$$E[\rho] = T_s[\rho] + \int d\mathbf{r} \left(V_{ext}(\mathbf{r}) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r} - \mathbf{R}_a|} \right) \rho(\mathbf{r}) + E_H[\rho] + E_{xc}[\rho]$$

Density from **non-interacting auxiliary system** that reproduces the **interacting density**

$$\rho(\mathbf{r}) = \sum_n f_n |\psi_n(\mathbf{r})|^2 \quad H_{KS}[\rho] |\psi_n\rangle = \varepsilon_n |\psi_n\rangle \quad H_{KS}[\rho] = \frac{\delta E}{\delta \rho}$$



DFT and qubits: valley-orbit coupling (VOC)



Conduction electrons in silicon have a wave function with support that may be spread across 6 “valleys”

$$\psi(\mathbf{r}) = \sum_{j=1}^6 F_j(\mathbf{r}) \phi_j(\mathbf{r})$$

Effective mass approximation - wave function factors
slowly varying envelope function $\times$ rapidly varying Bloch function

$$EF_l(\mathbf{r}) = \left(\hat{\mathbf{T}}_l + U(\mathbf{r}) \right) F_l(\mathbf{r}) + \sum_{j \in \pm\{x,y,z\}} V_{lj}^{VO}(\mathbf{r}) F_j(\mathbf{r})$$

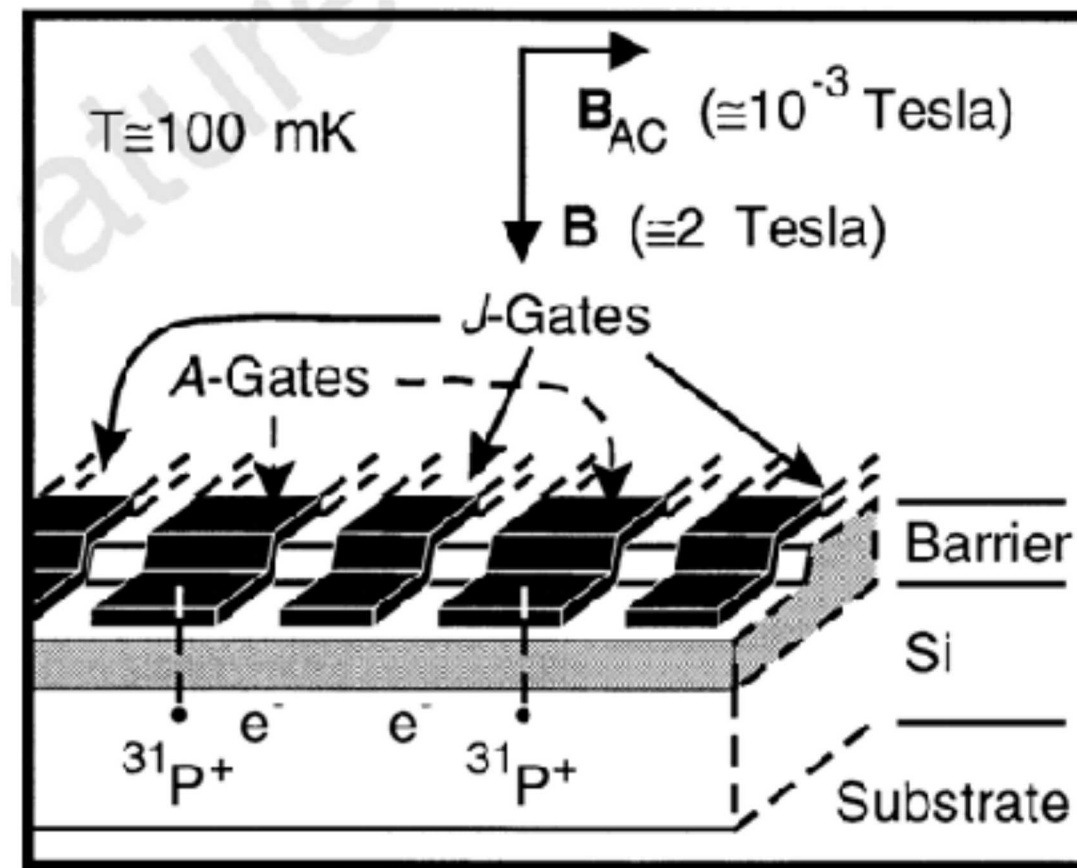
We can coarse grain and integrate out the Bloch functions,
except for a VOC term

Bloch functions
are computed
using DFT

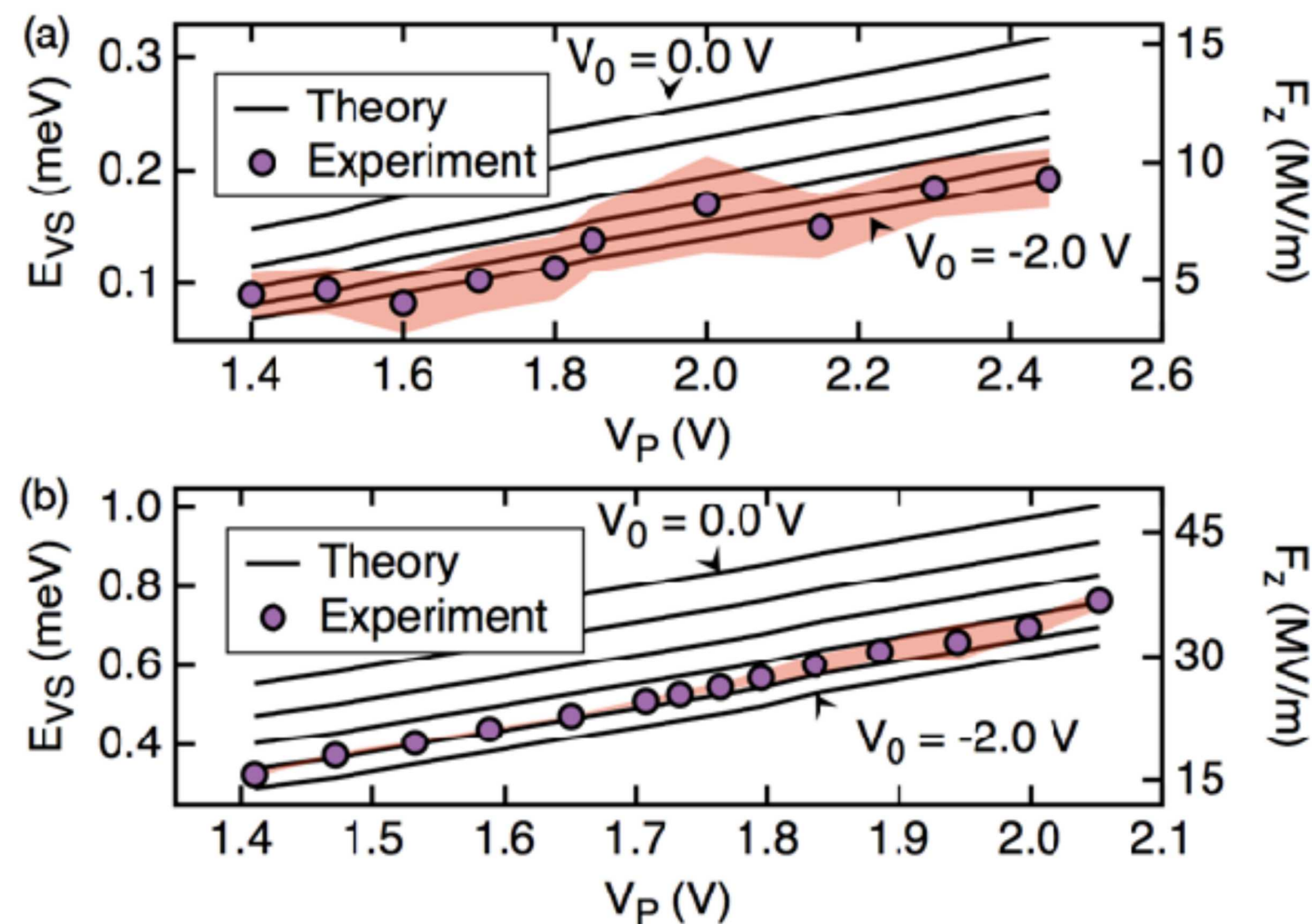
$$V_{lj}^{VO}(\mathbf{r}) \rightarrow \phi_l^*(\mathbf{r}) \phi_j(\mathbf{r}) U(\mathbf{r})$$

Largest when the potential varies on short length scales,
e.g., near shallow impurities and material interfaces

DFT and qubits: consequences of VOC



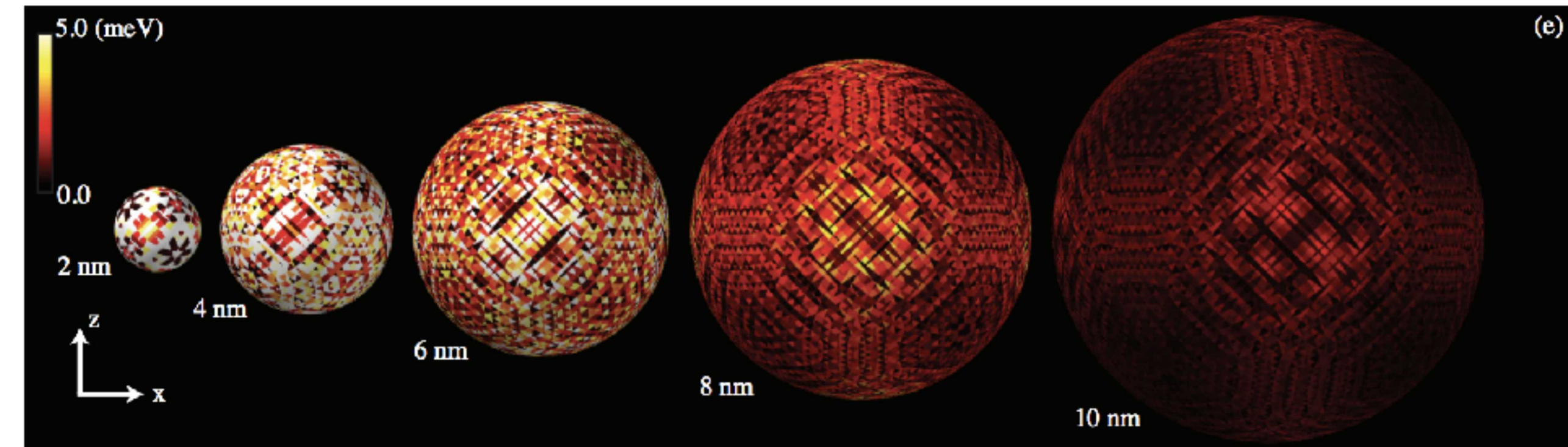
Kane, Nature (1998)



Gamble, et al., App. Phys. Lett. (2016)

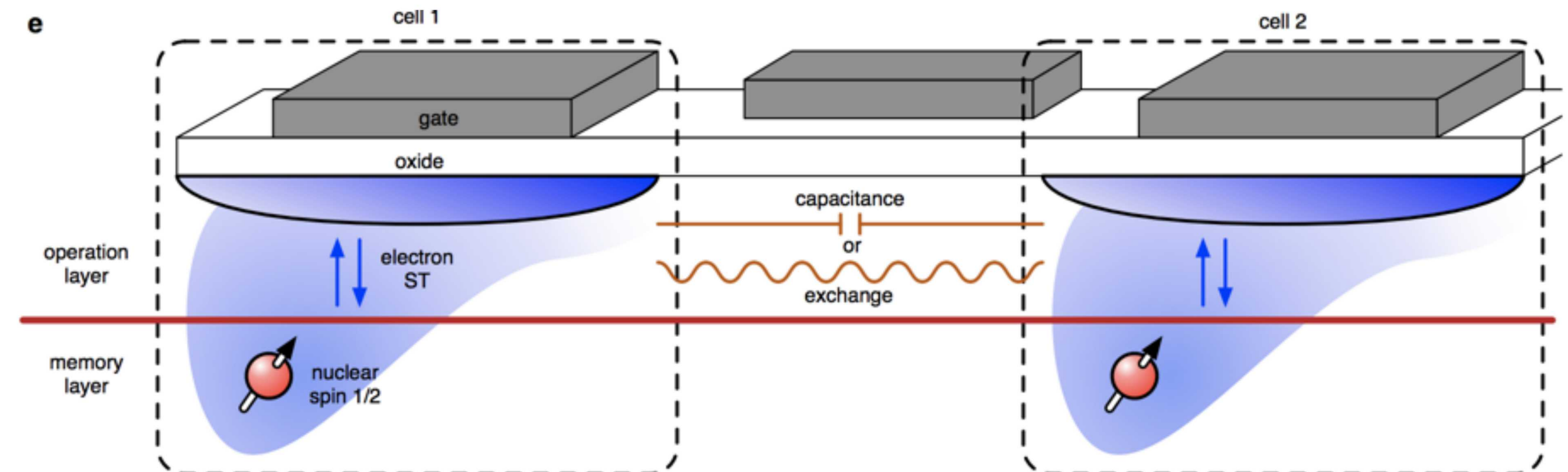
Multivalley effective mass theory simulation of donors in silicon

John King Gamble, N. Tobias Jacobson, Erik Nielsen, Andrew D. Baczewski, Jonathan E. Moussa, Inès Montañó, and Richard P. Muller
Phys. Rev. B **91**, 235318 – Published 29 June 2015



Coherent coupling between a quantum dot and a donor in silicon

Patrick Harvey-Collard, N. Tobias Jacobson, Martin Rudolph, Jason Dominguez, Gregory A. Ten Eyck, Joel R. Wendt, Tammy Pluym, John King Gamble, Michael P. Lilly, Michel Pioro-Ladrière & Malcolm S. Carroll

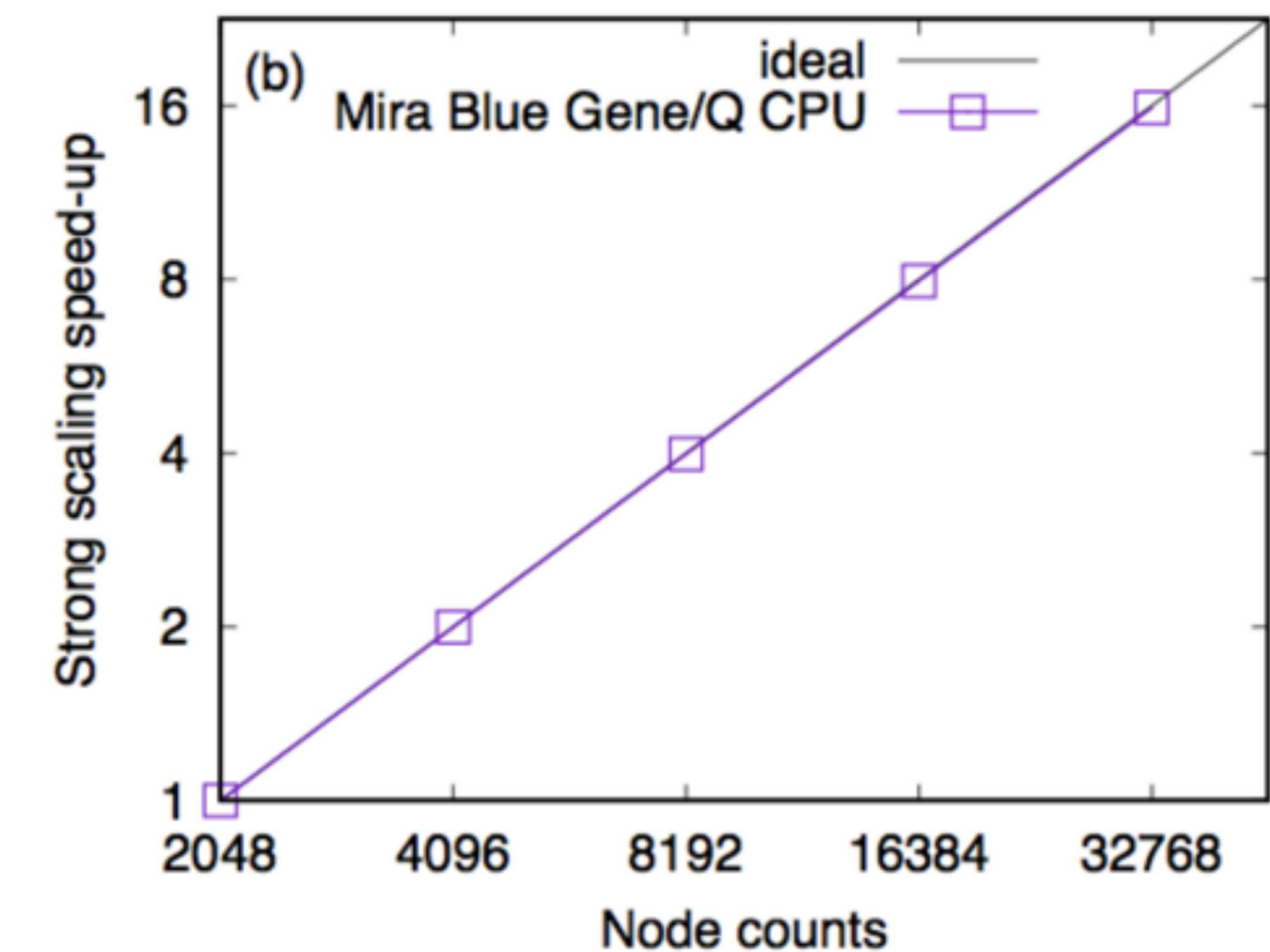
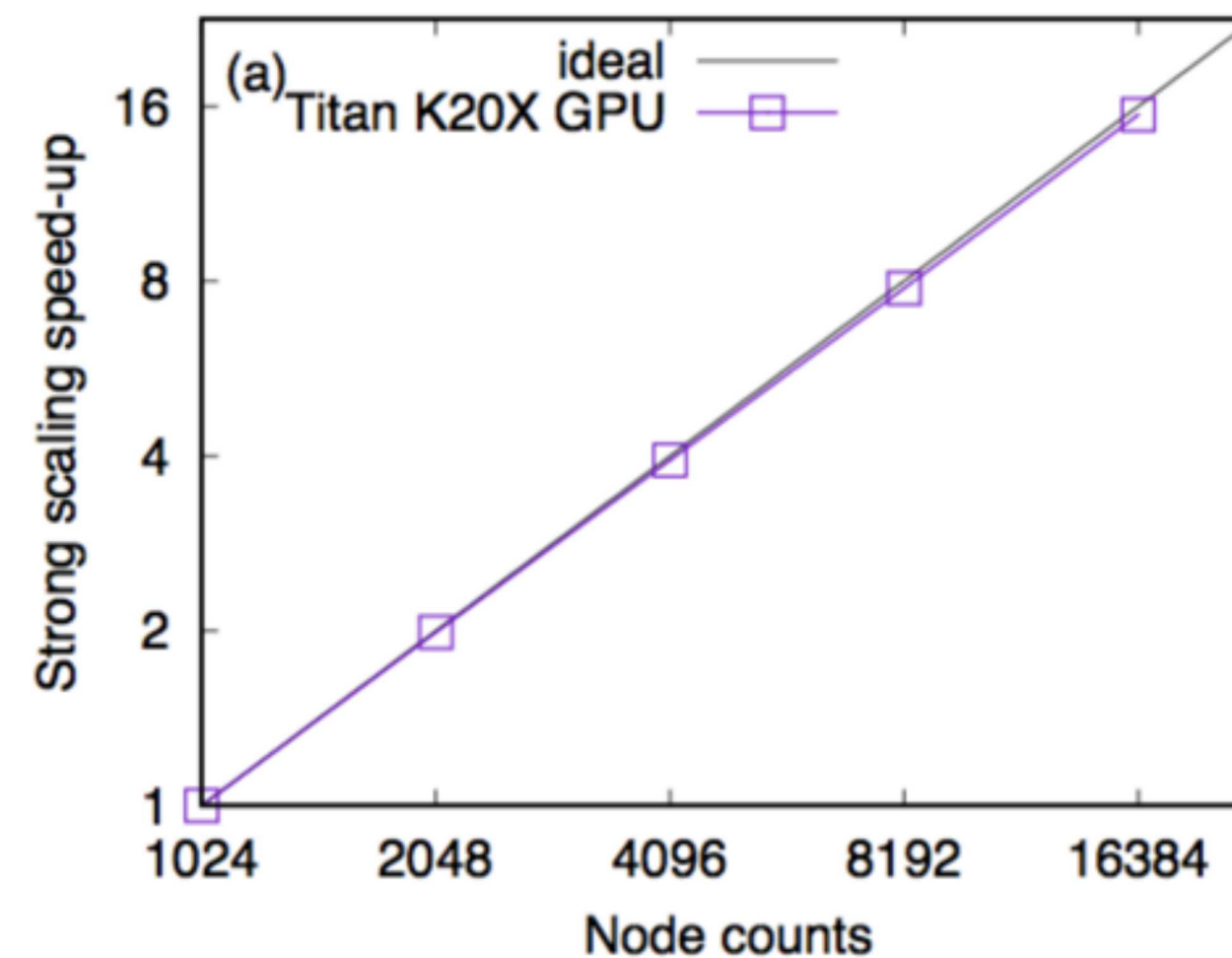


Quantum Monte Carlo (QMC)

QMC is becoming a more popular approach to materials simulation,
in part because it readily parallelizes onto *very large* HPC systems

QMCPACK: an open source *ab initio* quantum Monte Carlo package for the electronic structure of atoms, molecules and solids

Jeongnim Kim¹ , Andrew D Baczewski², Todd D Beaudet³, Anouar Benali^{4,5}, M Chandler Bennett⁶, Mark A Berrill⁷, Nick S Blunt⁸, Edgar Josué Landinez Borda⁹, Michele Casula¹⁰, David M Ceperley¹¹, Simone Chiesa¹¹, Bryan K Clark¹¹, Raymond C Clay III², Kris T Delaney¹², Mark Dewing⁵, Kenneth P Esler¹³, Hongxia Hao¹⁴, Olli Heinonen^{15,16}, Paul R C Kent^{17,18} , Jaron T Krogel¹⁹, Ilkka Kylänpää¹⁹, Ying Wai Li²⁰, M Graham Lopez⁷, Ye Luo^{4,5} , Fionn D Malone⁹ , Richard M Martin¹¹, Amrita Mathuriya¹, Jeremy McMinis⁹, Cody A Melton⁶, Lubos Mitas⁶, Miguel A Morales⁹, Eric Neuscamman^{21,22} , William D Parker²³ , Sergio D Pineda Flores²¹, Nichols A Romero^{4,5}, Brenda M Rubenstein¹⁴, Jacqueline A R Shea²¹, Hyeondeok Shin⁵, Luke Shulenburger², Andreas F Tillack²⁰, Joshua P Townsend² , Norm M Tubman²¹, Brett Van Der Goetz²¹, Jordan E Vincent¹¹, D ChangMo Yang²⁴ , Yubo Yang¹¹, Shuai Zhang⁹ and Luning Zhao²¹ — [Hide full author list](#)



Pros:

- Does not break the [variational principle](#)
- Dilemma: “efficient & biased” or “inefficient & unbiased”
- Works directly with a representation of the [full many-body wave function](#) or [Green’s function](#)

Cons:

- State-of-the-art QMC studies can eat [10s of millions of CPU hours](#) for just a few data points
- Different manifestations of [fermionic sign problem](#), ultimately NP-Complete (*Troyer & Wiese, PRL (2005)*)
- Treatment of [core states](#) is still challenging

QMC in a bit more detail

$$H = \sum_{i=1}^{N_e} \left(-\frac{\nabla_i^2}{2} + V_{ext}(\mathbf{r}_i) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} \right) + \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Uses **Monte Carlo integration** to evaluate energies for a particular wave function ansatz

$$E_{\Psi_T} = \frac{\langle \Psi_T | H | \Psi_T \rangle}{\langle \Psi_T | \Psi_T \rangle}$$

Slater-Jastrow ansatz is a popular choice for going beyond DFT* $\Psi_T = D^\uparrow D^\downarrow e^J$

Projector (or diffusion) Monte Carlo = **imaginary time phase estimation**

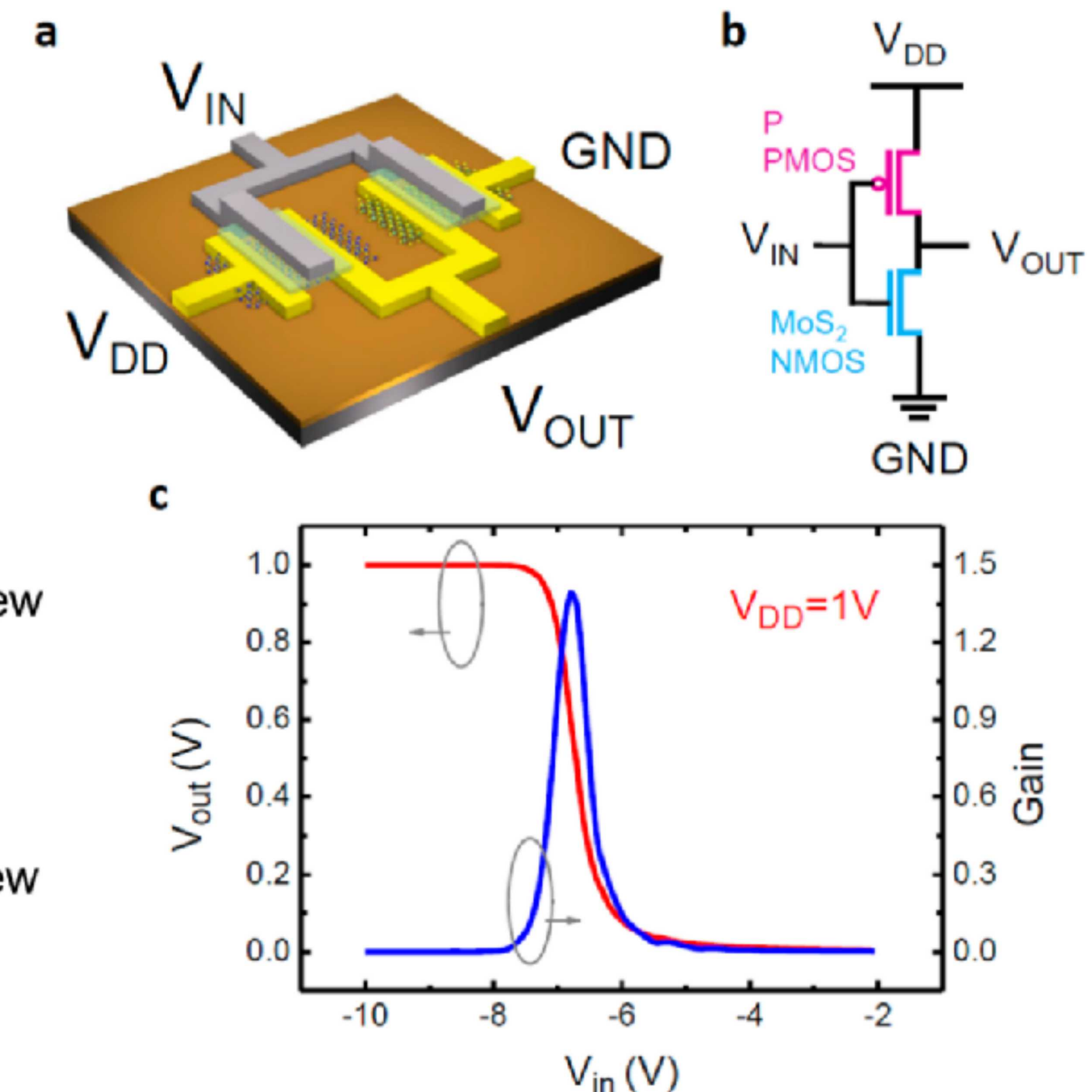
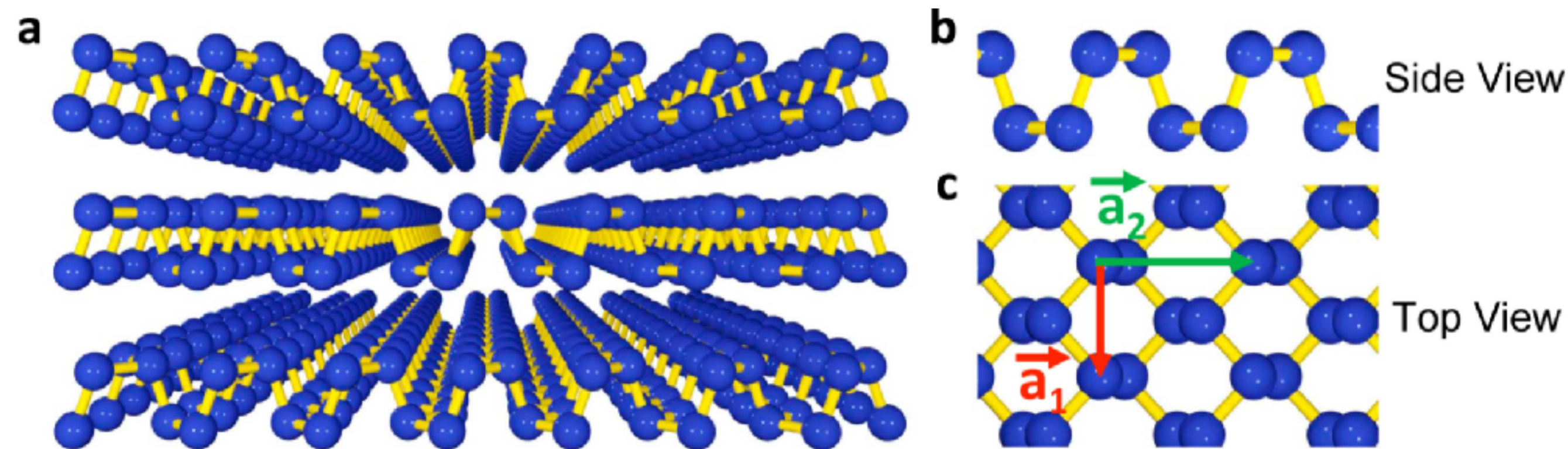
$$\lim_{\beta \rightarrow \infty} G(\beta) |\Psi_T\rangle = e^{-\beta E_0} |\Psi_T\rangle \quad G(\beta) = e^{-\beta H}$$

*Multi-determinant Slater-Jastrow or auxiliary field QMC are growing in popularity

(Nearly) 2D Materials

Phosphorene: An Unexplored 2D Semiconductor with a High Hole Mobility

Han Liu,^{†,‡} Adam T. Neal,^{†,‡} Zhen Zhu,[§] Zhe Luo,^{‡,⊥} Xianfan Xu,^{‡,⊥} David Tománek,[§] and Peide D. Ye^{†,‡,*}



Few-layer phosphorene is nominally held together by the
van der Waals' (vdW) interaction

vdW physics is manifestly non-local and challenging to capture using DFT

QMC vs DFT

NANO LETTERS

Letter

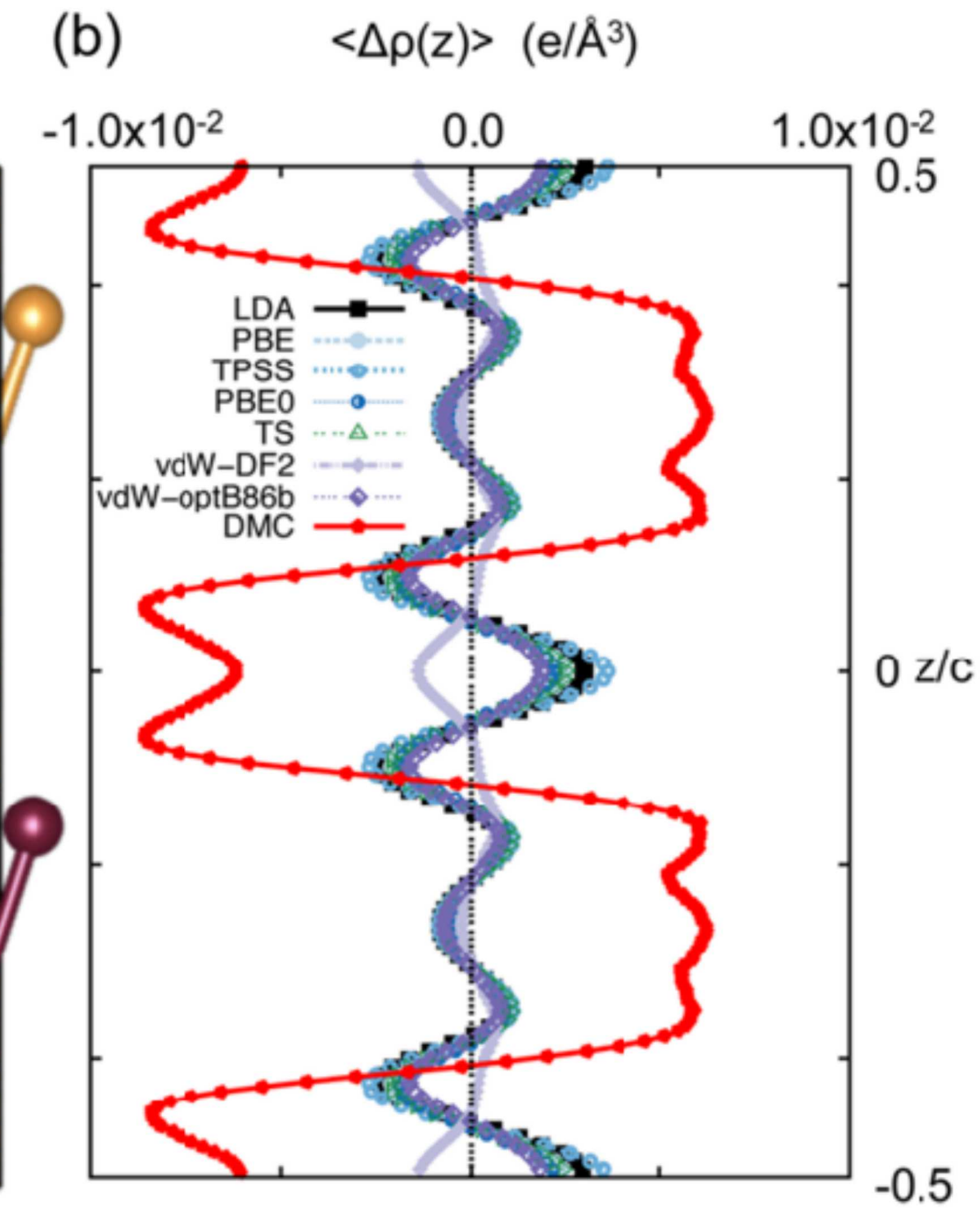
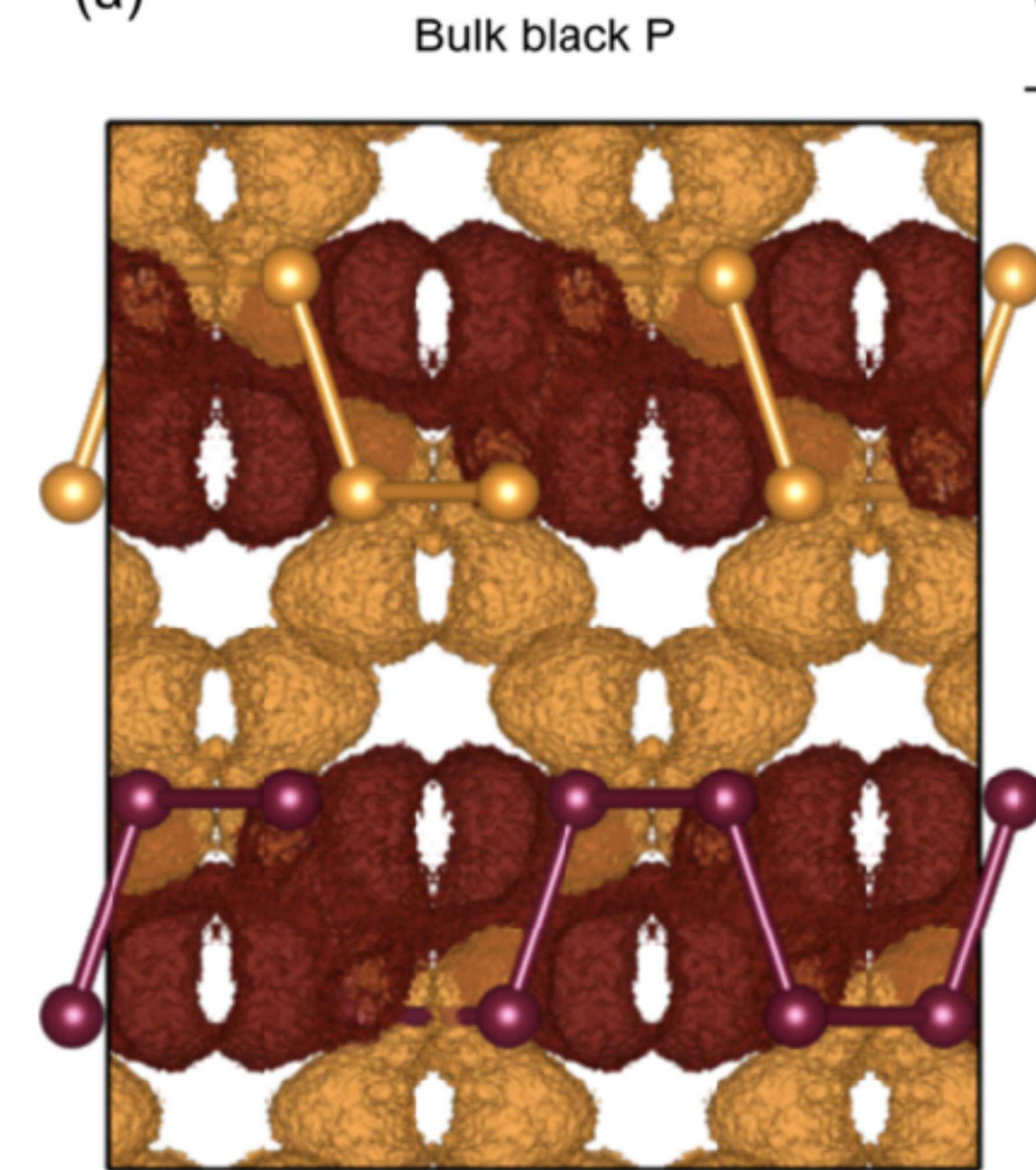
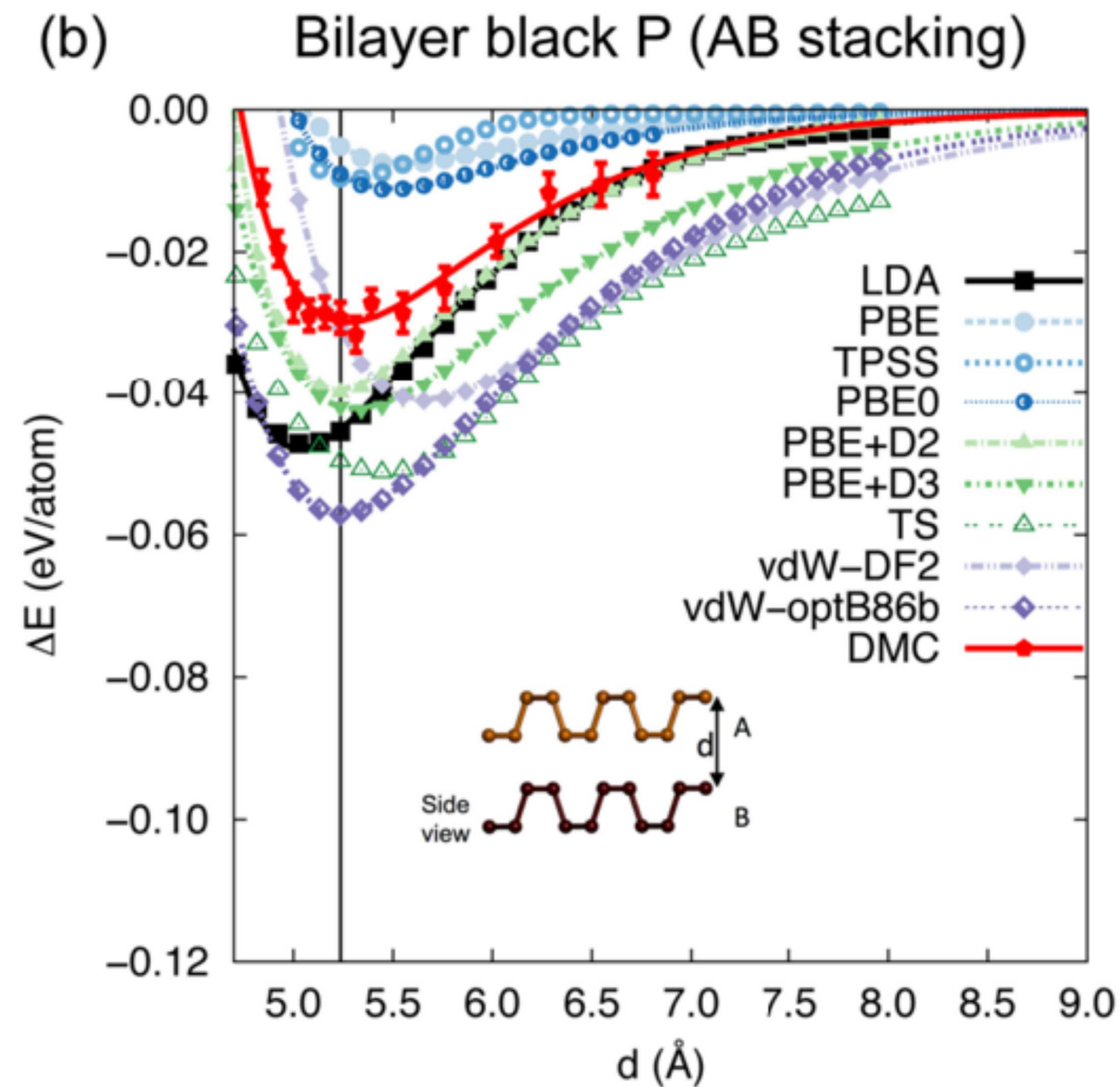
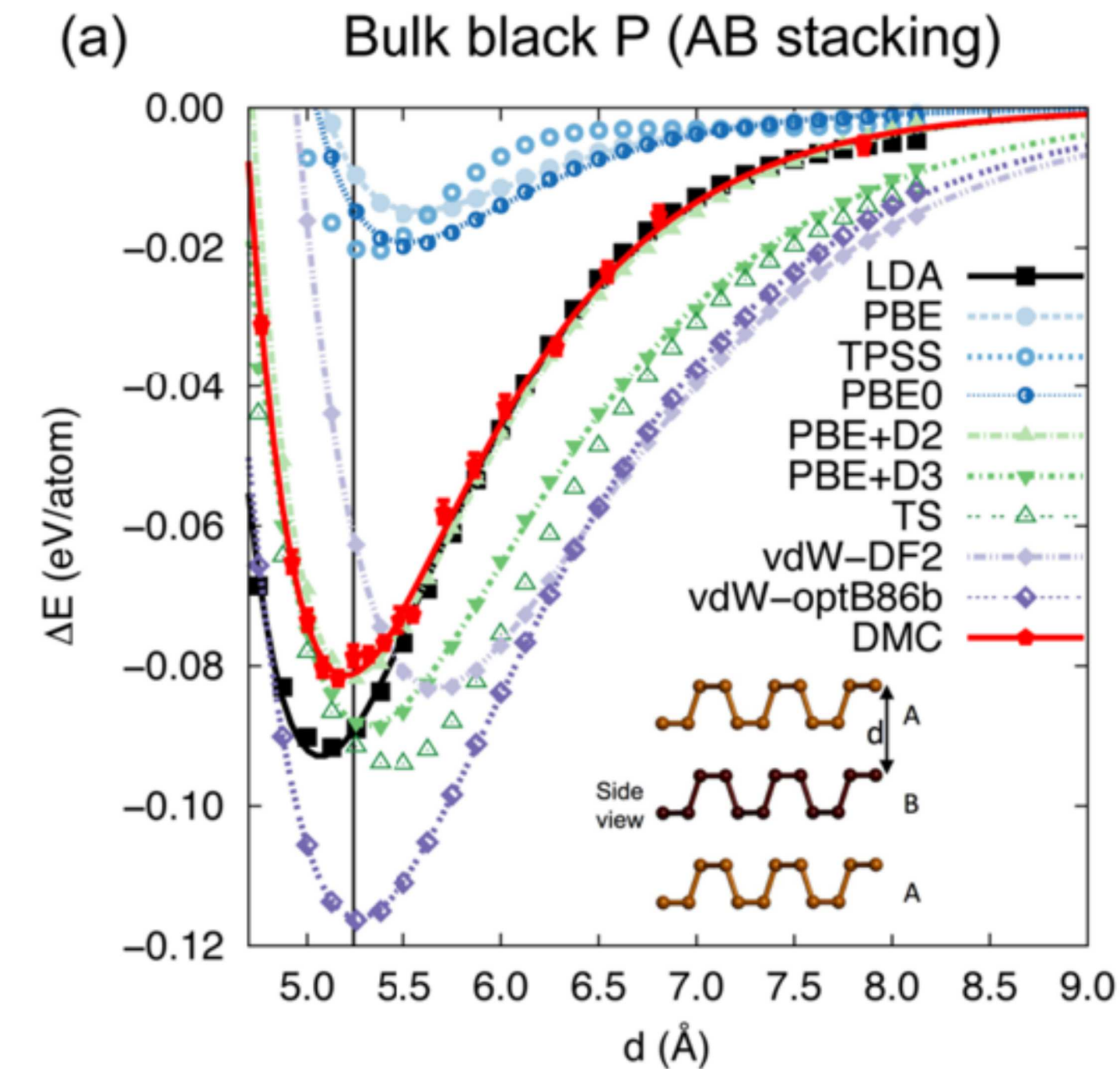
pubs.acs.org/NanoLett

The Nature of the Interlayer Interaction in Bulk and Few-Layer Phosphorus

L. Shulenburger,[†] A.D. Baczewski,[‡] Z. Zhu,[‡] J. Guan,[‡] and D. Tománek

(a)

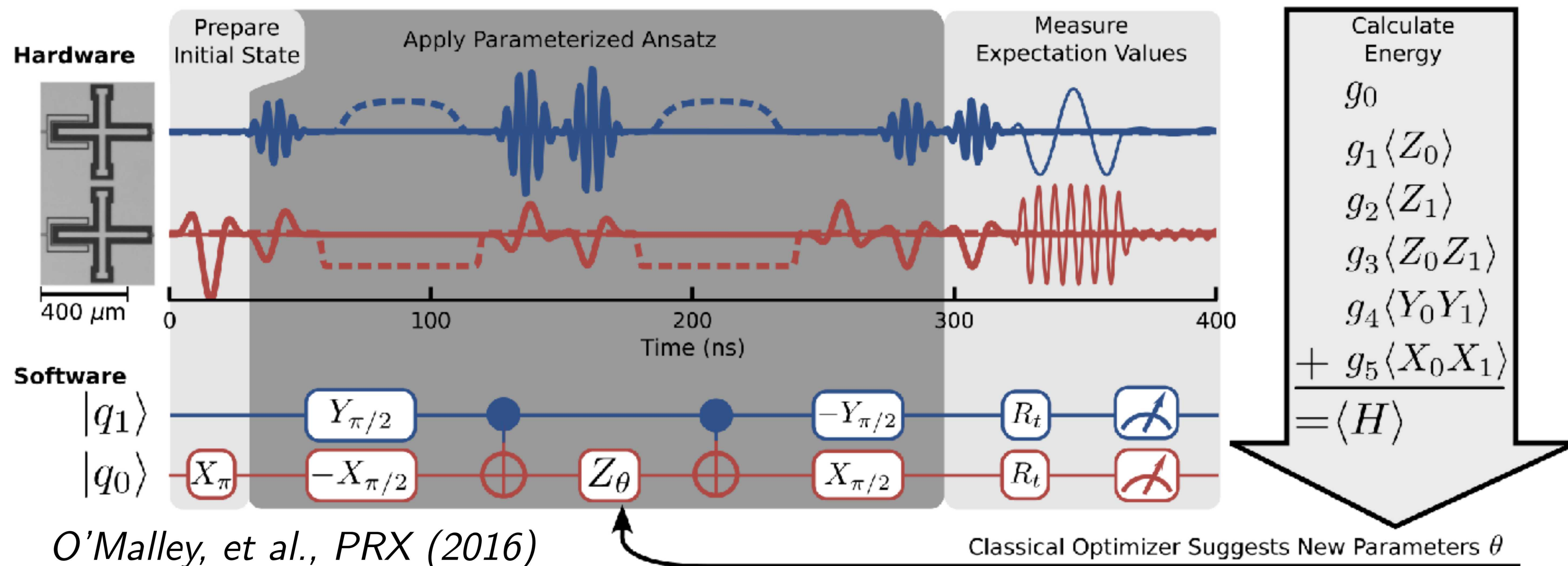
(b)



Least sophisticated DFT functional does the best,
All of them fail to capture interlayer bonding qualitatively
Error cancellation = right answer for the wrong reason!

Error cancellation in quantum simulation

The **wrong Hamiltonian** might still give us the **right answer**



DFT: total energy = weighted sum of local(ish) observables

VQE: total energy = weighted sum of low-weight measurements

Classical heuristics may give insight into the robustness of simulation on NISQ hardware

Time-dependent DFT (TDDFT)

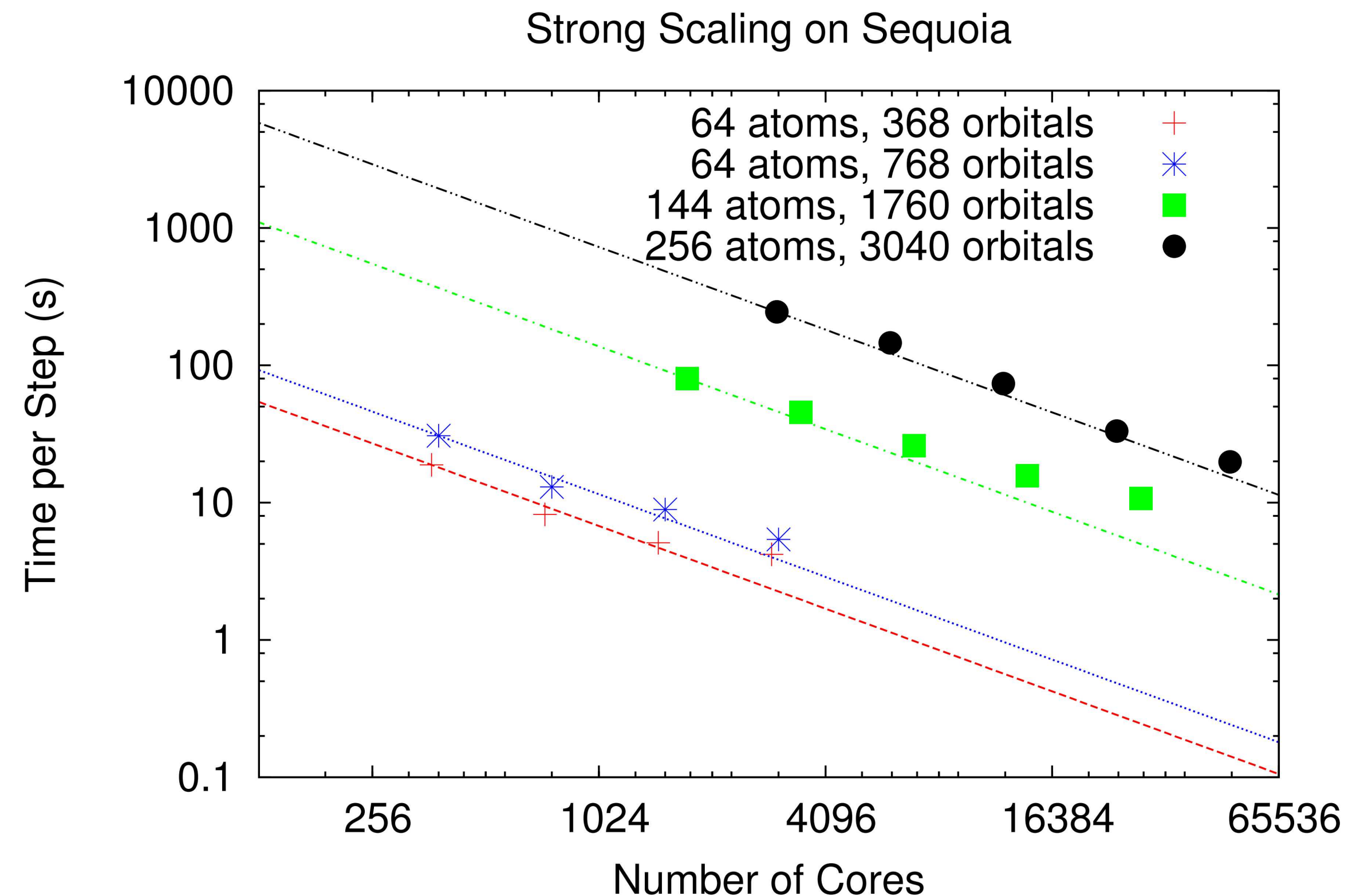
TDDFT upgrades DFT from a ground state theory to an excited state theory

Pros:

- Non-perturbative, requires exchange-correlation functional
- Linear-scaling with system size, typically decides cost vs. accuracy dilemma
- Very scalable like QMC, but less accurate
- Unlike DFT, TDDFT is in BQP!
(Whitfield, et al., New J. Phys. (2014))

Cons:

- Non-perturbative, requires exchange-correlation functional
- The exact exchange-correlation functional time non-local
- Tends to have steps and other difficult to describe features

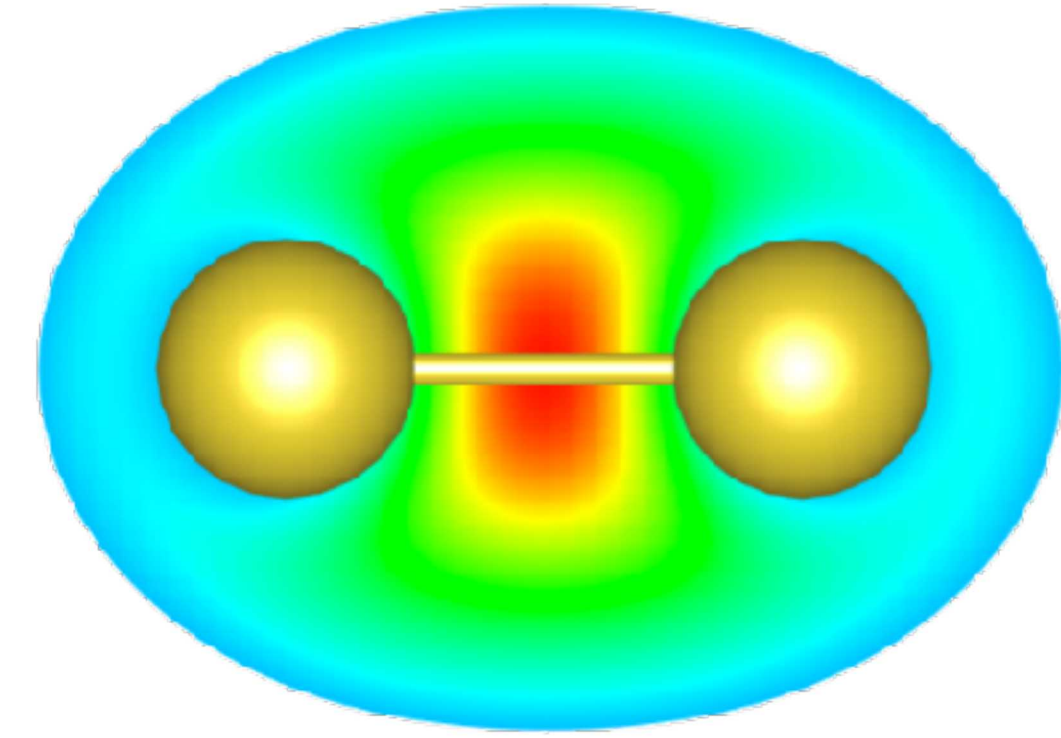


TDDFT in a bit more detail

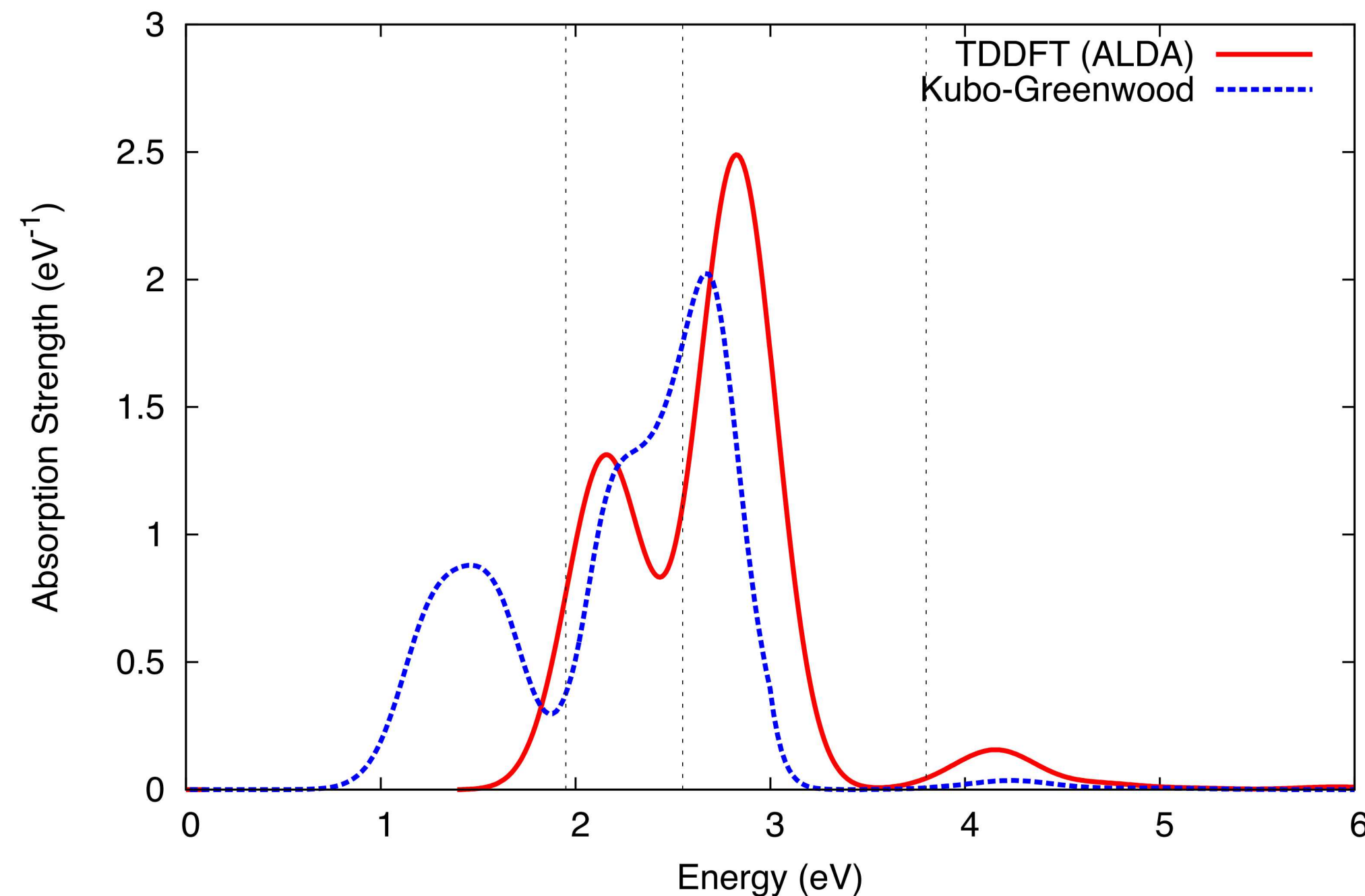
Start from a ground state DFT calculation and integrate time-dependent Kohn-Sham equations

$$H_{KS}[\rho] |\psi_n\rangle = \varepsilon_n |\psi_n\rangle$$

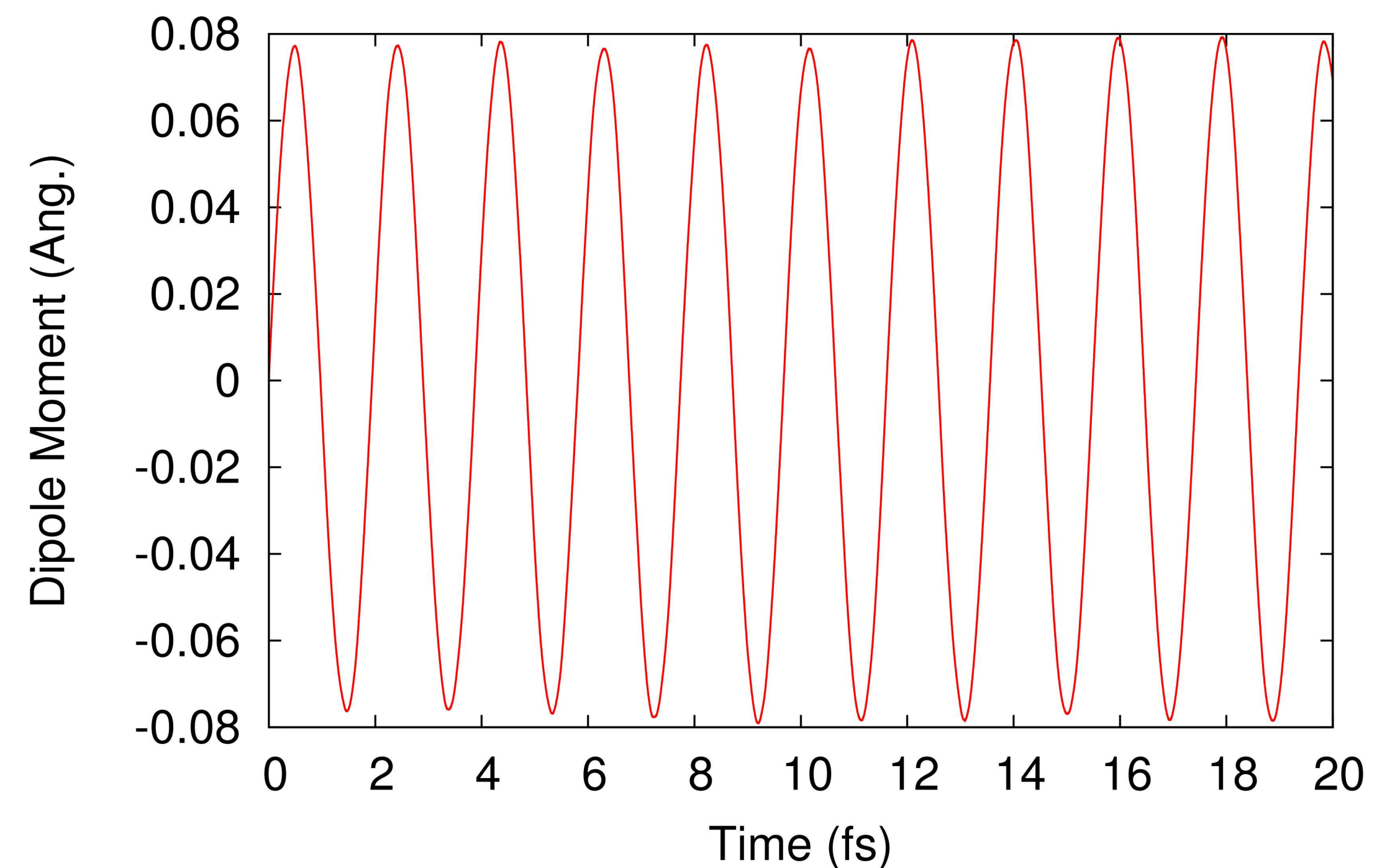
$$i \frac{d}{dt} |\psi_n\rangle = H_{KS}[\rho(\mathbf{r}, t), t] |\psi_n\rangle$$



Dipole Response of a Sodium Dimer



Response to Longitudinal Excitation



X-ray Thomson Scattering (XRTS) Basics

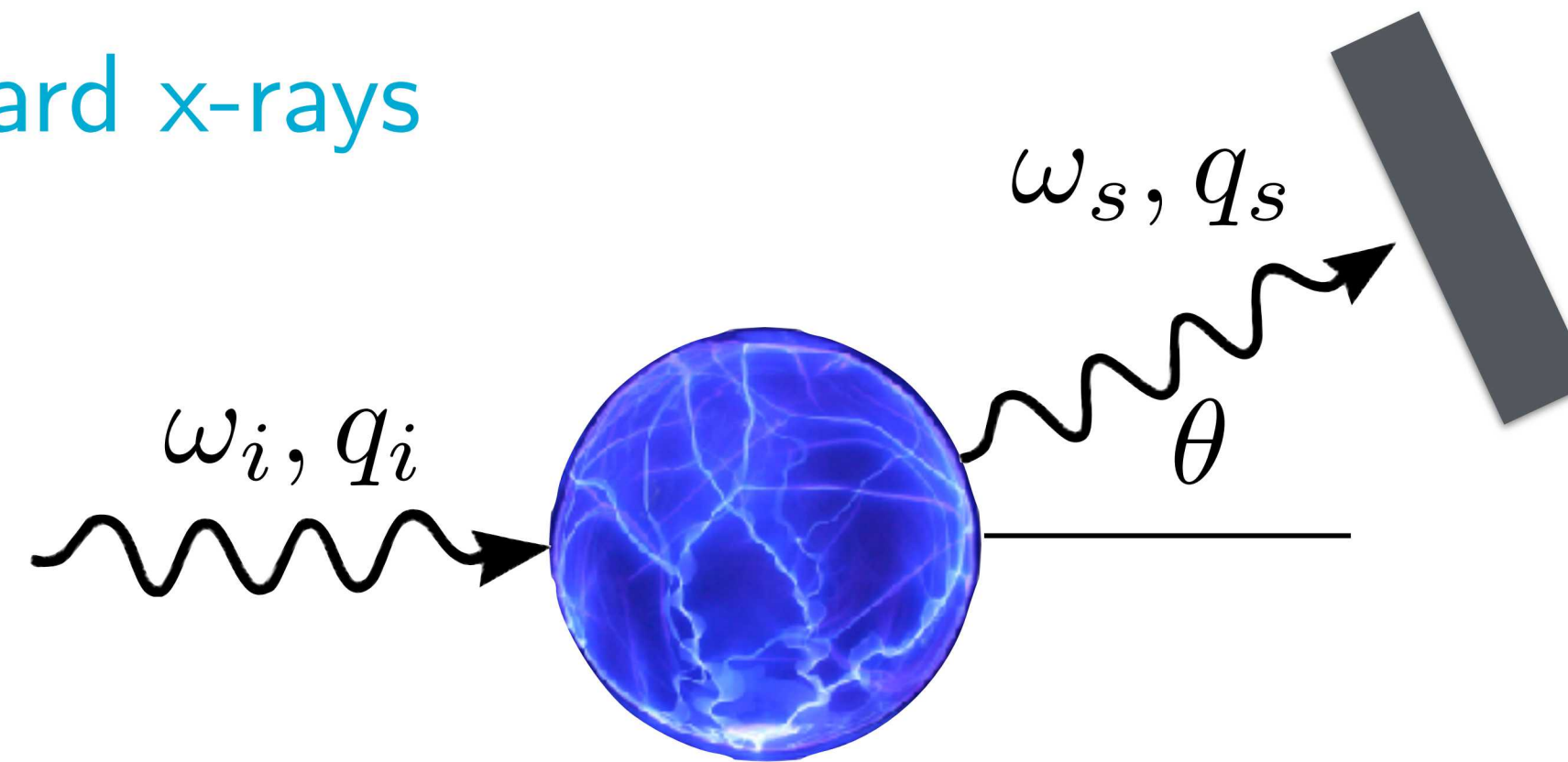
X-ray Thomson scattering in high energy density plasmas

Siegfried H. Glenzer and Ronald Redmer

Rev. Mod. Phys. **81**, 1625 – Published 1 December 2009

Measure **inelastically scattered x-rays**

Penetrate with **hard x-rays**



$$\begin{aligned}\omega &= \omega_i - \omega_s \\ q &= q_i - q_s\end{aligned}$$

$$q = 2q_i \sin(\theta/2)$$

Sample of WDM

(opaque to optical probes)

$$\frac{d^2\sigma}{d\Omega d\omega} = \sigma_T \frac{q_s}{q_i} S(q, \omega)$$

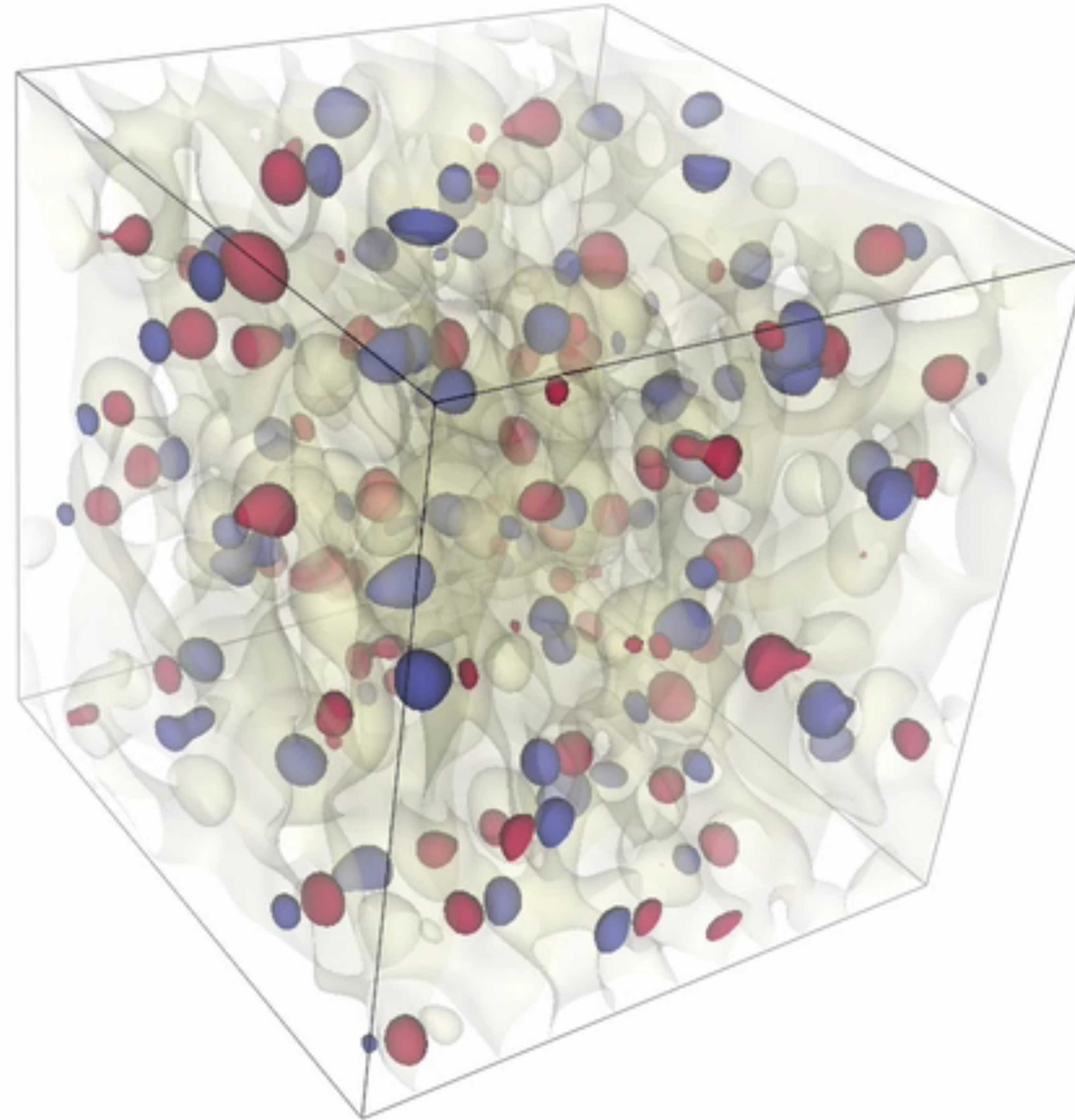
Cross section proportional to **dynamic structure factor (DSF)**

Probes **density, ionization state, structure, temperature, and...**

TDDFT for XRTS

X-ray Thomson Scattering in Warm Dense Matter without the Chihara Decomposition

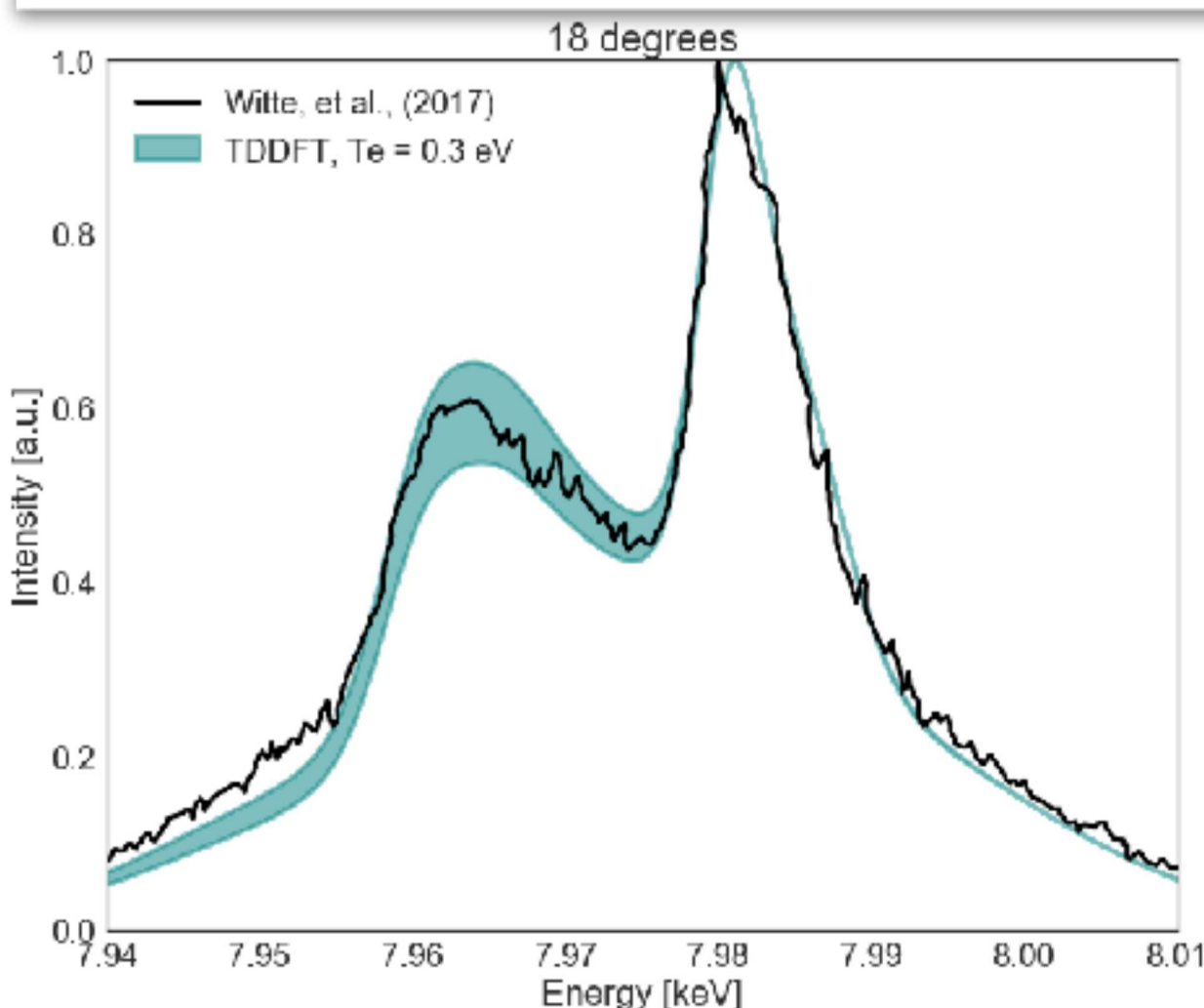
A. D. Baczewski, L. Shulenburger, M. P. Desjarlais, S. B. Hansen, and R. J. Magyar
Phys. Rev. Lett. **116**, 115004 – Published 18 March 2016



TDDFT constrains experiments

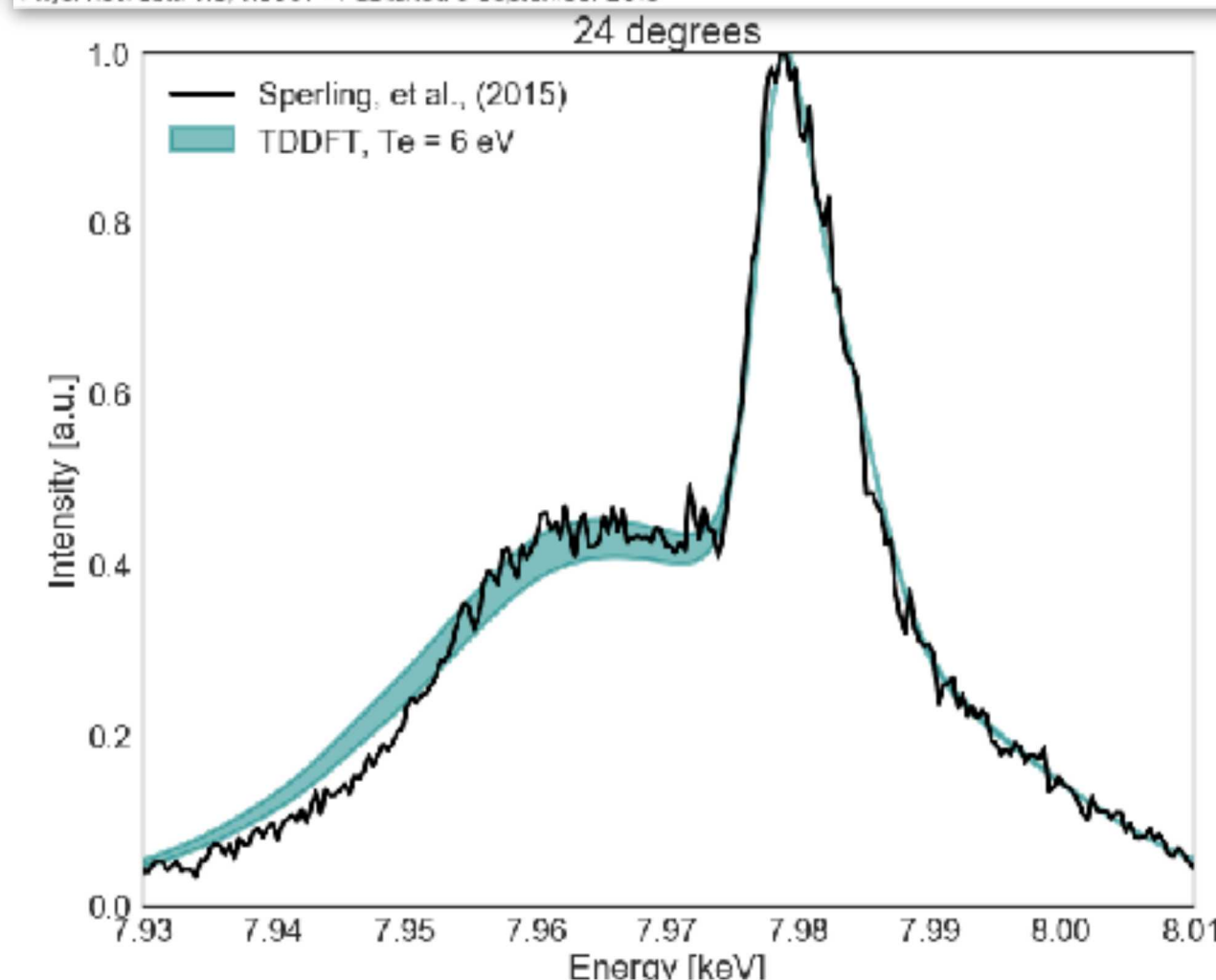
Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

B. B. L. Witte, L. B. Fletcher, E. Galtier, E. Gamboa, H. J. Lee, U. Zastrau, R. Redmer, S. H. Glenzer, and P. Sperling
Phys. Rev. Lett. **118**, 225001 – Published 31 May 2017



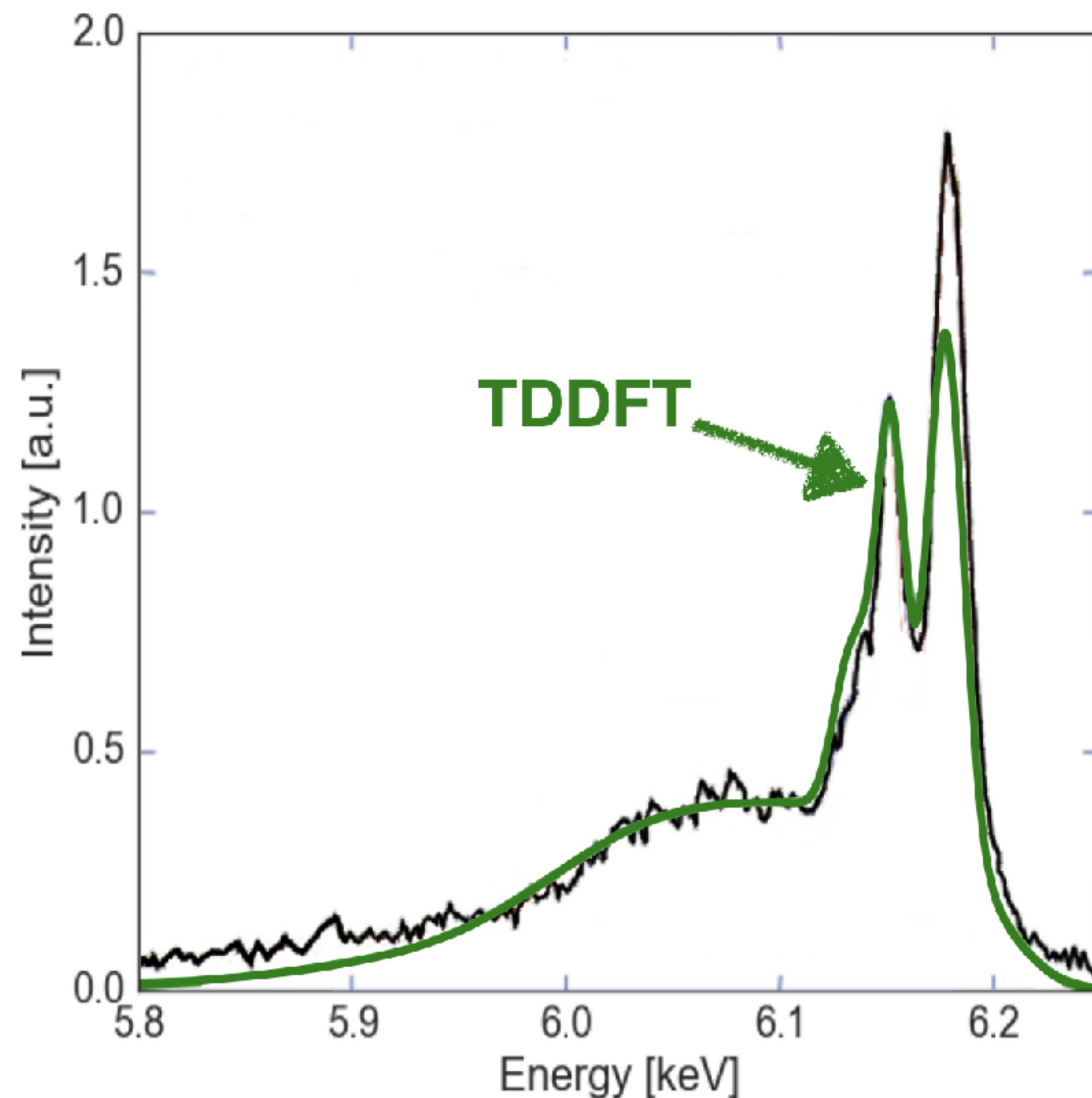
Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakayeva, H. Reinholz, G. Röpke, U. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer
Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015



X-Ray Thomson-Scattering Measurements of Density and Temperature in Shock-Compressed Beryllium

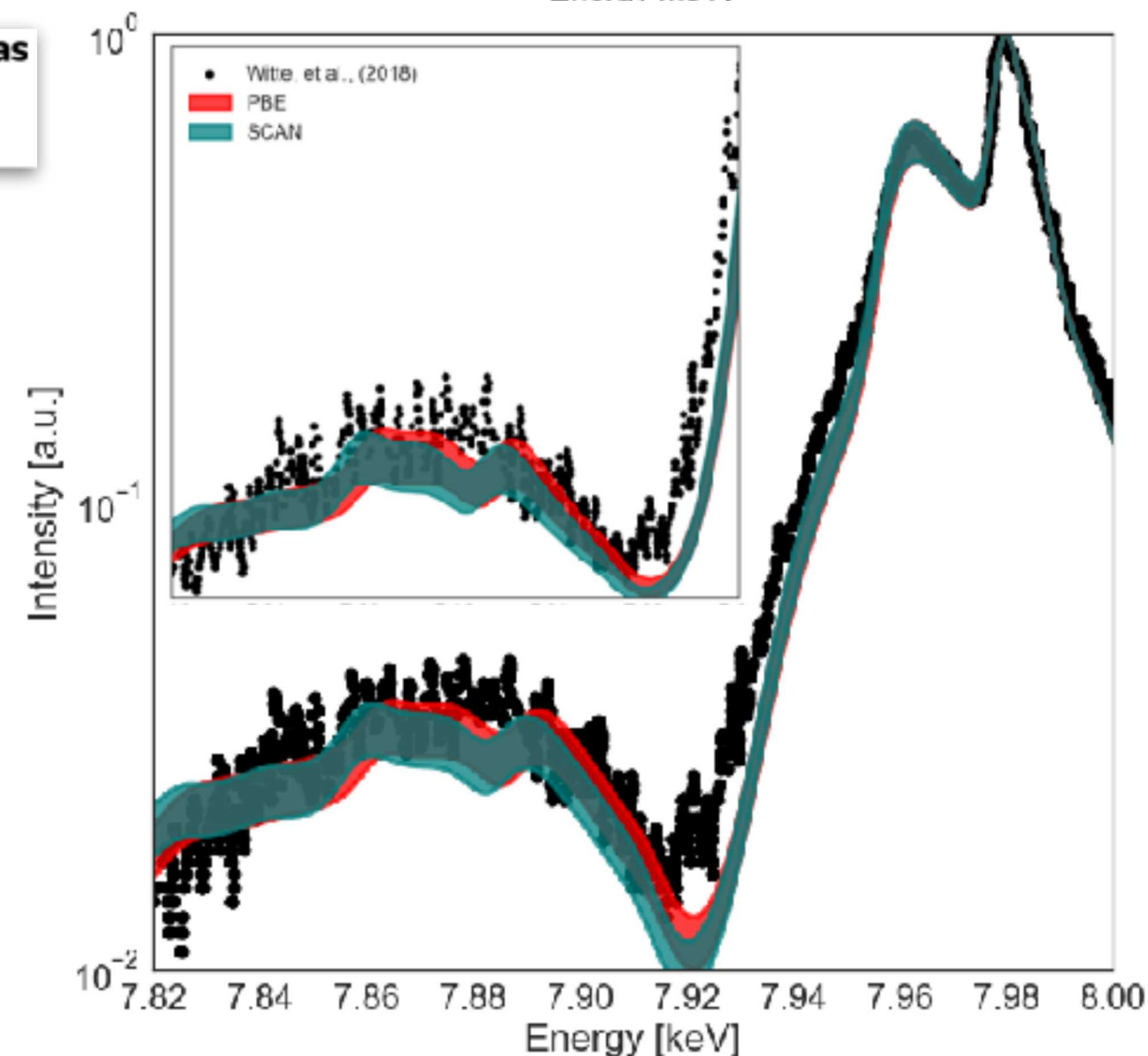
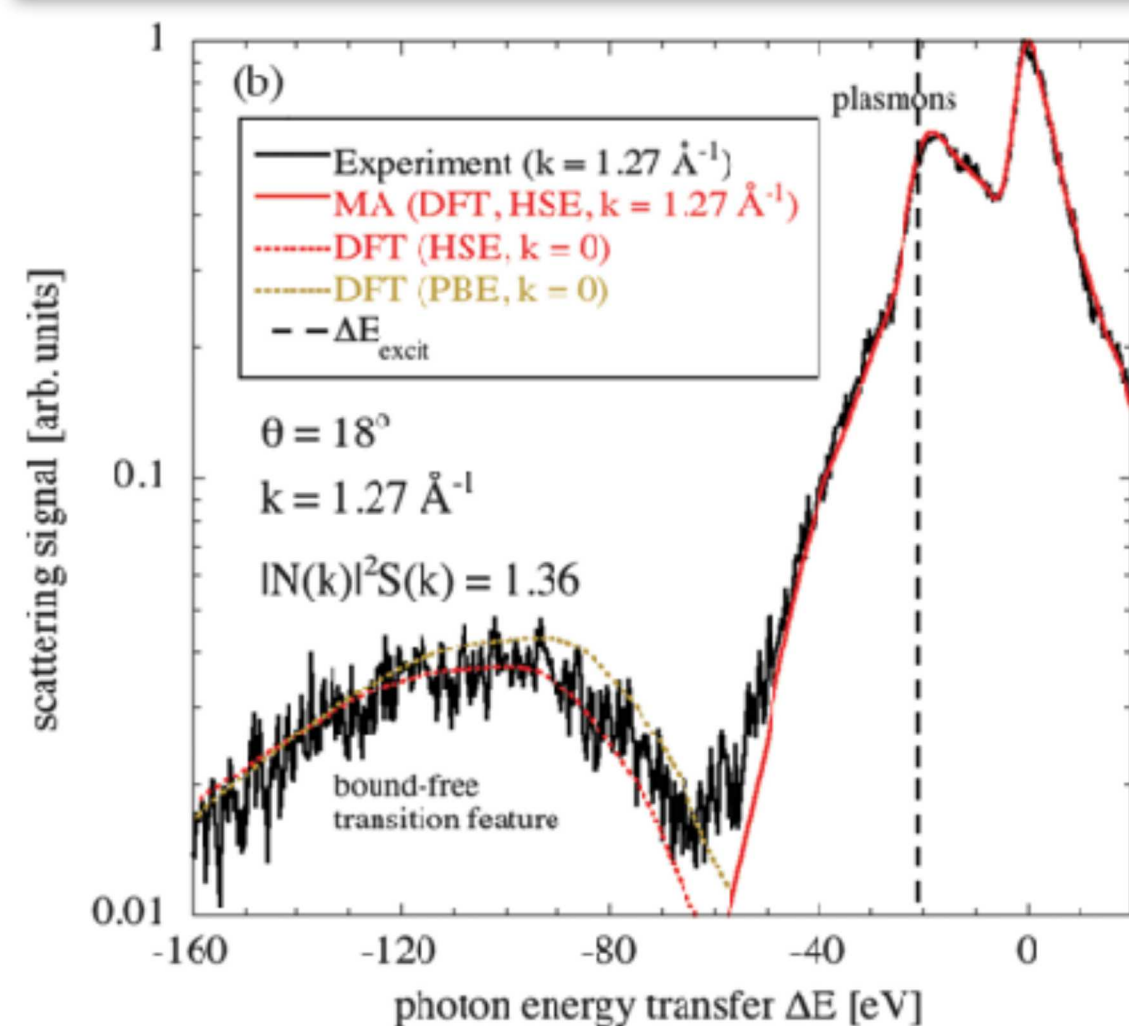
H. J. Lee, P. Neumayer, J. Castor, T. Döppner, R. W. Falcone, C. Fortmann, B. A. Hammel, A. L. Kitcher, O. L. Landen, R. W. Lee, D. D. Meyerhofer, D. H. Munro, R. Redmer, S. P. Regan, S. Weber, and S. H. Glenzer
Phys. Rev. Lett. **102**, 115001 – Published 16 March 2009



Observations of non-linear plasmon damping in dense plasmas

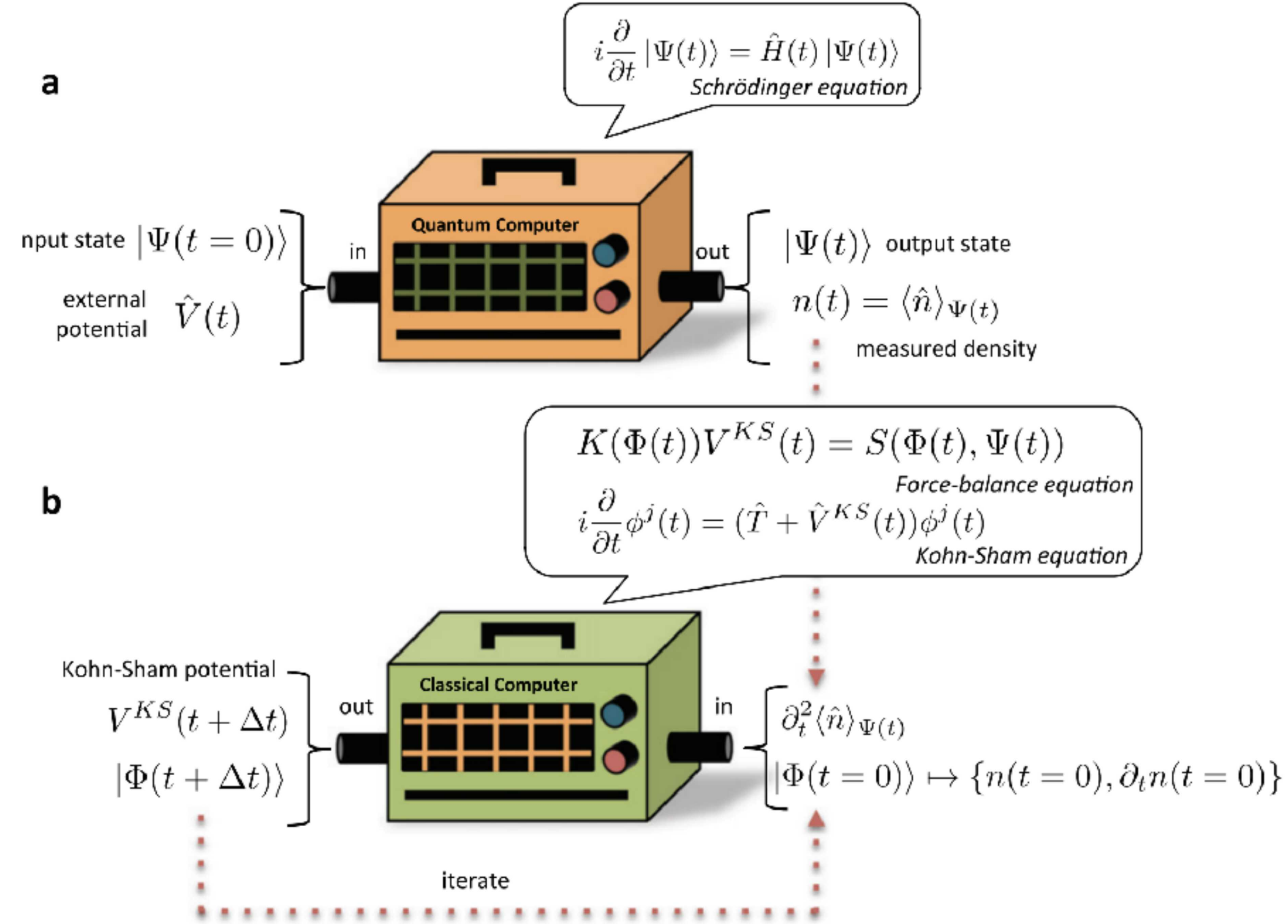
Physics of Plasmas **25**, 056901 (2018); <https://doi.org/10.1063/1.5317689>

B. B. L. Witte^{1,2,*}, P. Sperling^{1,2,3}, M. French⁴, V. Recaules⁵, S. H. Glenzer¹, and R. Redmer²



Quantum potential inversion

I mentioned that TDDFT is in BQP...



Whitfield et al., New J. Phys. (2014)

Quantum hardware might help us make TDDFT better

Coupled cluster theory

$$H = \sum_{i=1}^{N_e} \left(-\frac{\nabla_i^2}{2} + V_{ext}(\mathbf{r}_i) - \sum_{a=1}^{N_n} \frac{Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} \right) + \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Post-Hartree-Fock* theory: wave function ansatz is generated by exponential acting on a Hartree-Fock (or other) reference state $|\Psi\rangle = e^T |\Phi_{HF}\rangle$

Nominally **systematically improvable** $T = T_1 + T_2 + \dots$

$$T_n = \frac{1}{(n!)^2} \sum_{o_1, o_2, \dots, o_n} \sum_{u_1, u_2, \dots, u_n} t_{o_1, o_2, \dots, o_n}^{u_1, u_2, \dots, u_n} \hat{a}_{u_1}^\dagger \hat{a}_{u_2}^\dagger \dots \hat{a}_{u_n}^\dagger \hat{a}_{o_1} \hat{a}_{o_2} \dots \hat{a}_{o_n}$$

The generator for the ansatz is a **non-unitary operator**, we could use an **unitary ansatz** but this is extremely inefficient on classical hardware

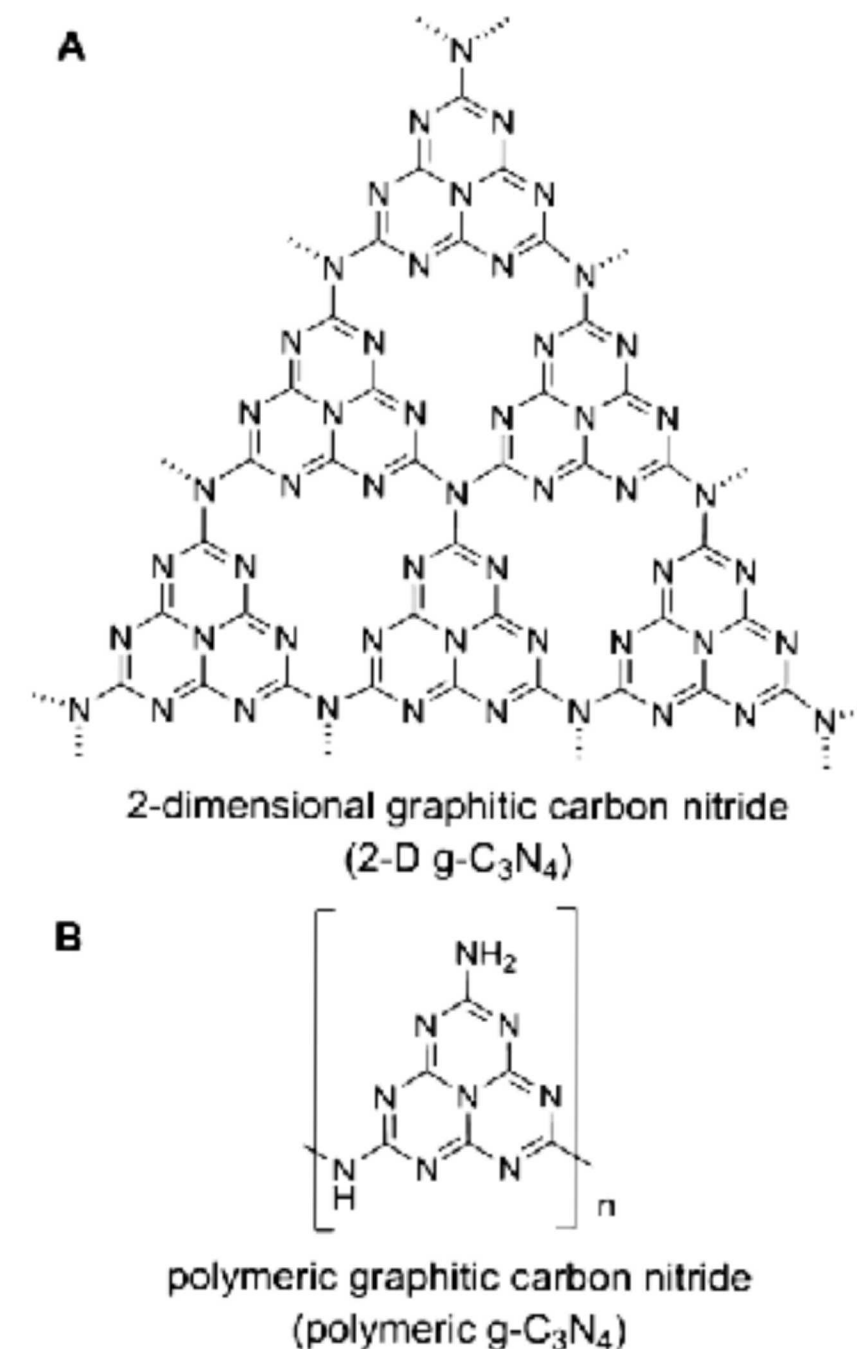
*Hartree-Fock generates a single Slater determinant that minimizes the expectation value of H

(Photo)catalysis

One of the vaunted applications of quantum simulation technologies (see FeMoCo).

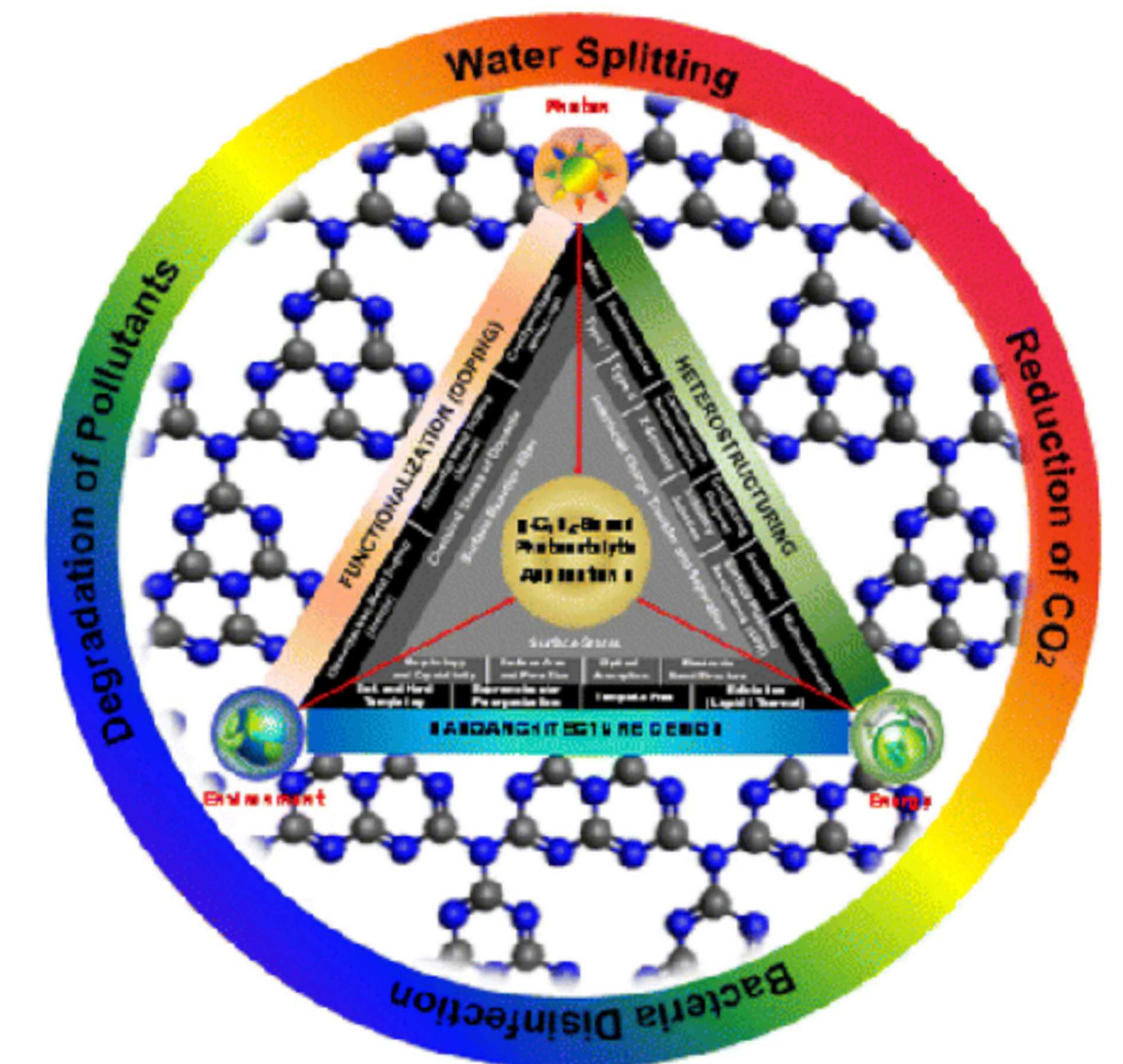
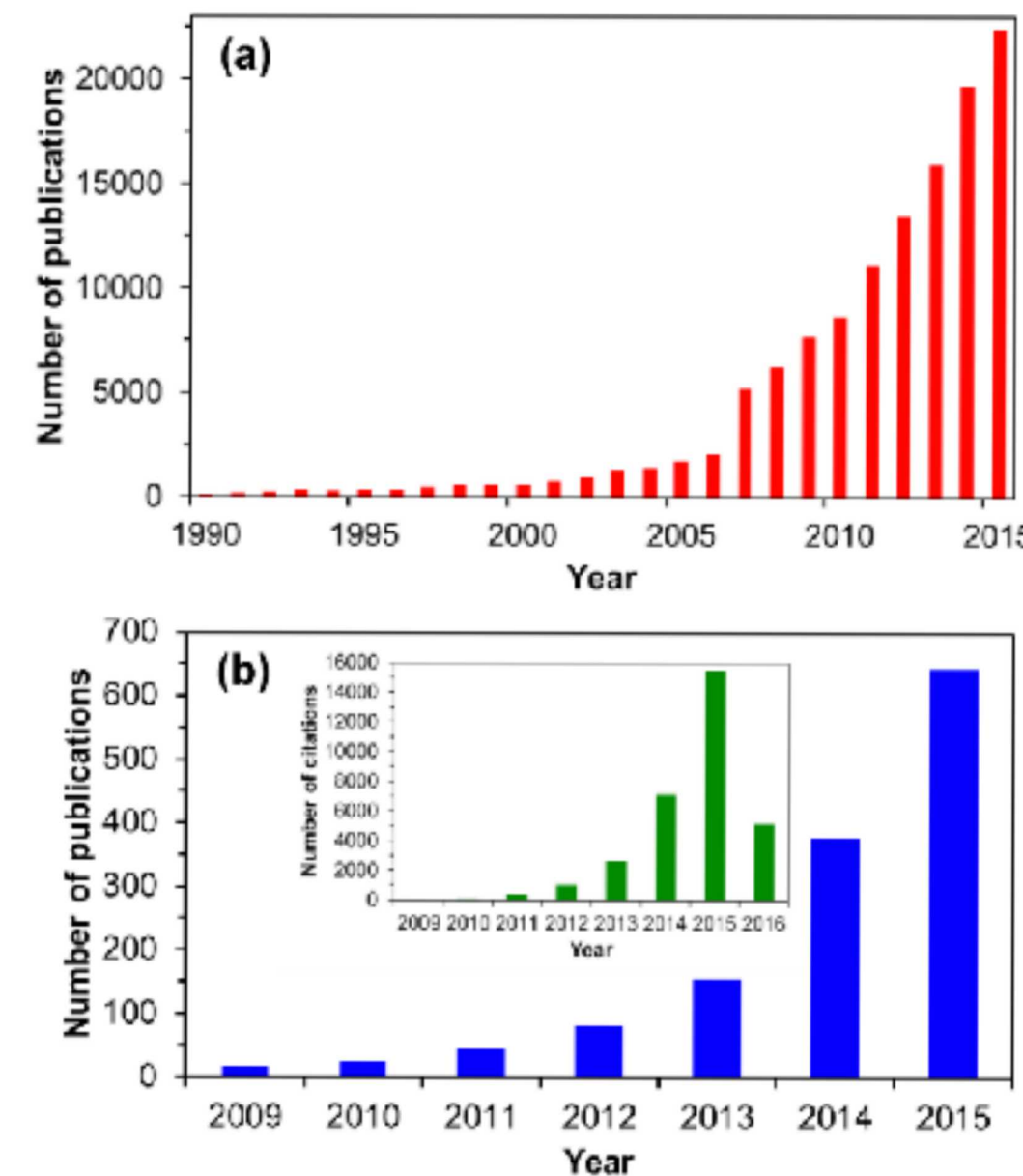
Large calculations that require the resolution of tiny energy differences.

Modeling photocatalysis adds the challenge of representing excited states



Graphitic Carbon Nitride ($g-C_3N_4$)-Based Photocatalysts for Artificial Photosynthesis and Environmental Remediation: Are We a Step Closer To Achieving Sustainability?

Wee-Jun Ong,^{*,†} Lling-Ling Tan,[†] Yun Hau Ng,[§] Siek-Ting Yong,[†] and Siang-Piao Chai^{*,†}

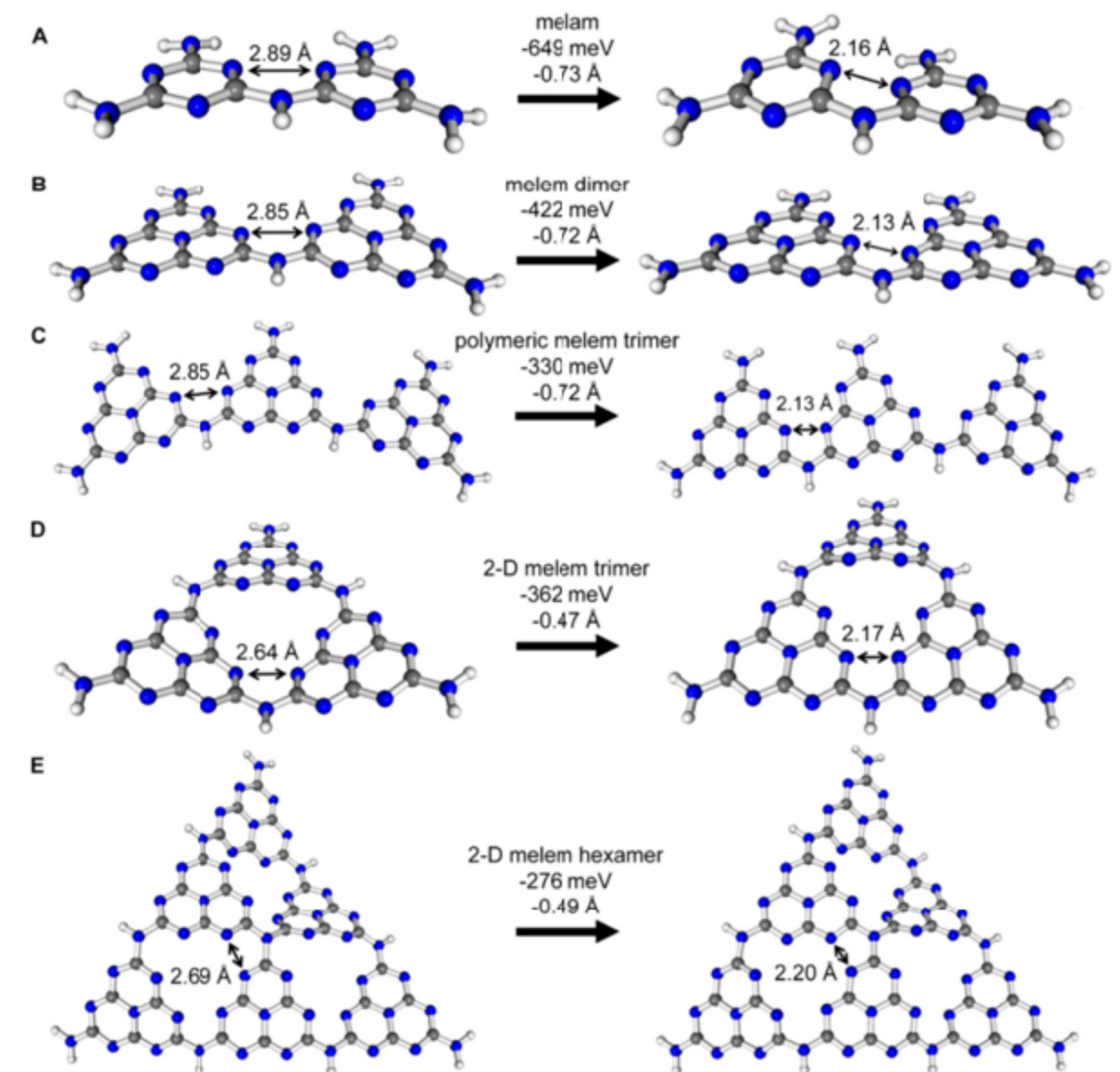
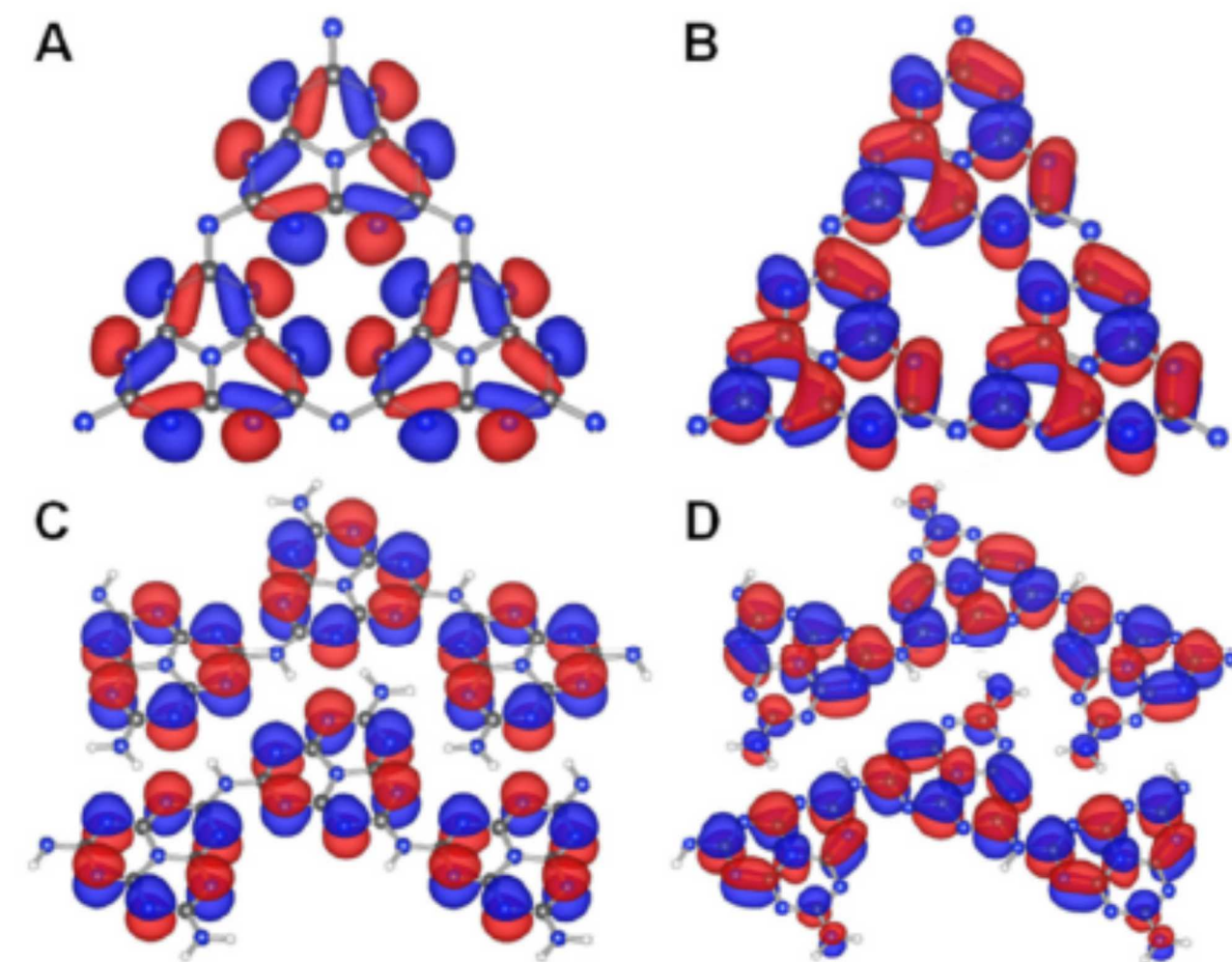
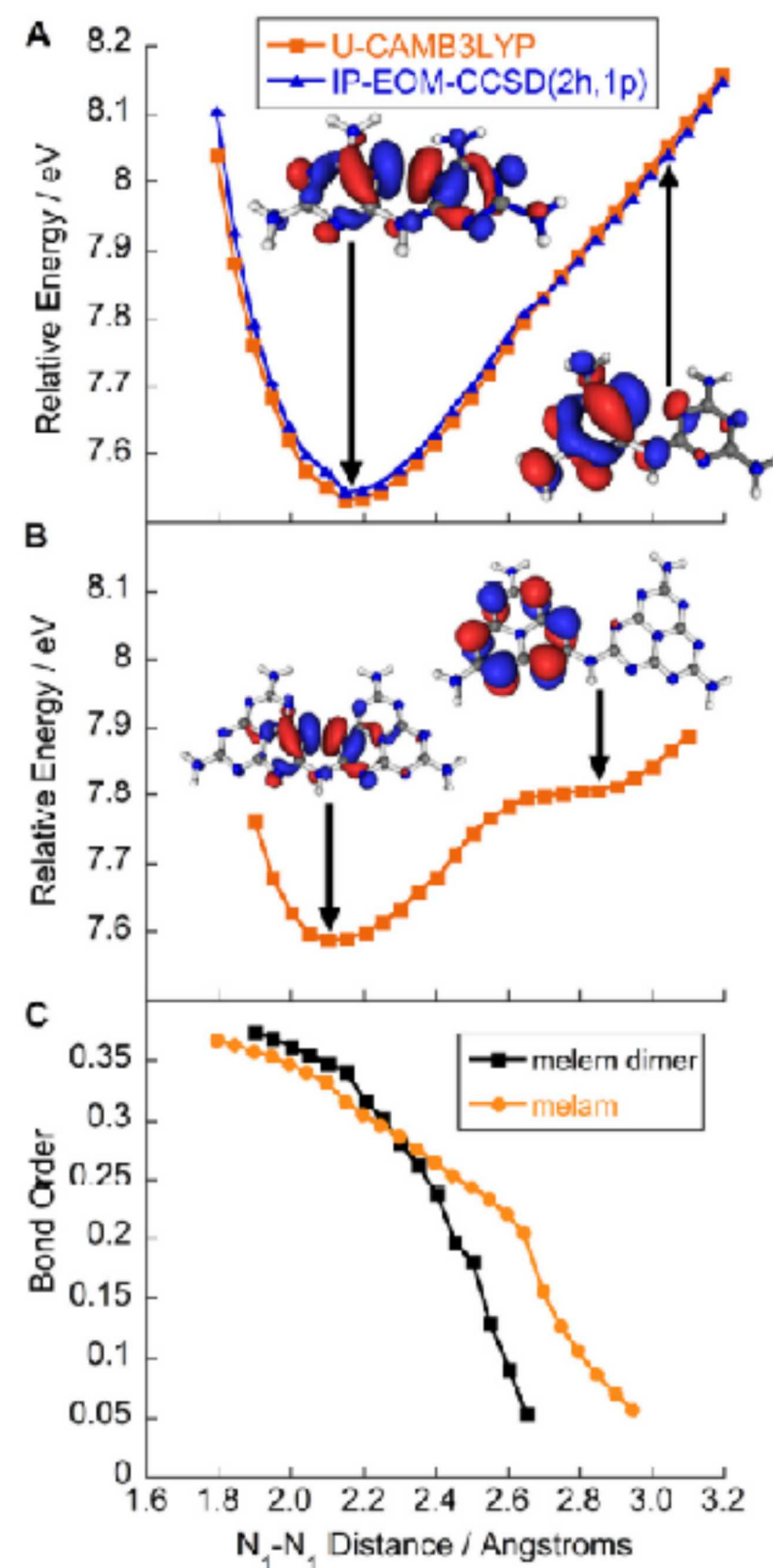


Photocatalysis and g-CNs

To understand hydrogen evolution, we must need higher levels of theory - right?

Cationic radicals of melem subunits responsible for catalytic action

DFT does a surprisingly good job!



We can be confident in going to bigger systems with DFT

Unitary CC for VQE

Non-unitary ansatz for **classical HW**

$$|\Psi\rangle = e^T |\Phi_{HF}\rangle$$

Unitary ansatz for **NISQ HW**

$$|\Psi\rangle = e^{T-T^\dagger} |\Phi_{HF}\rangle$$

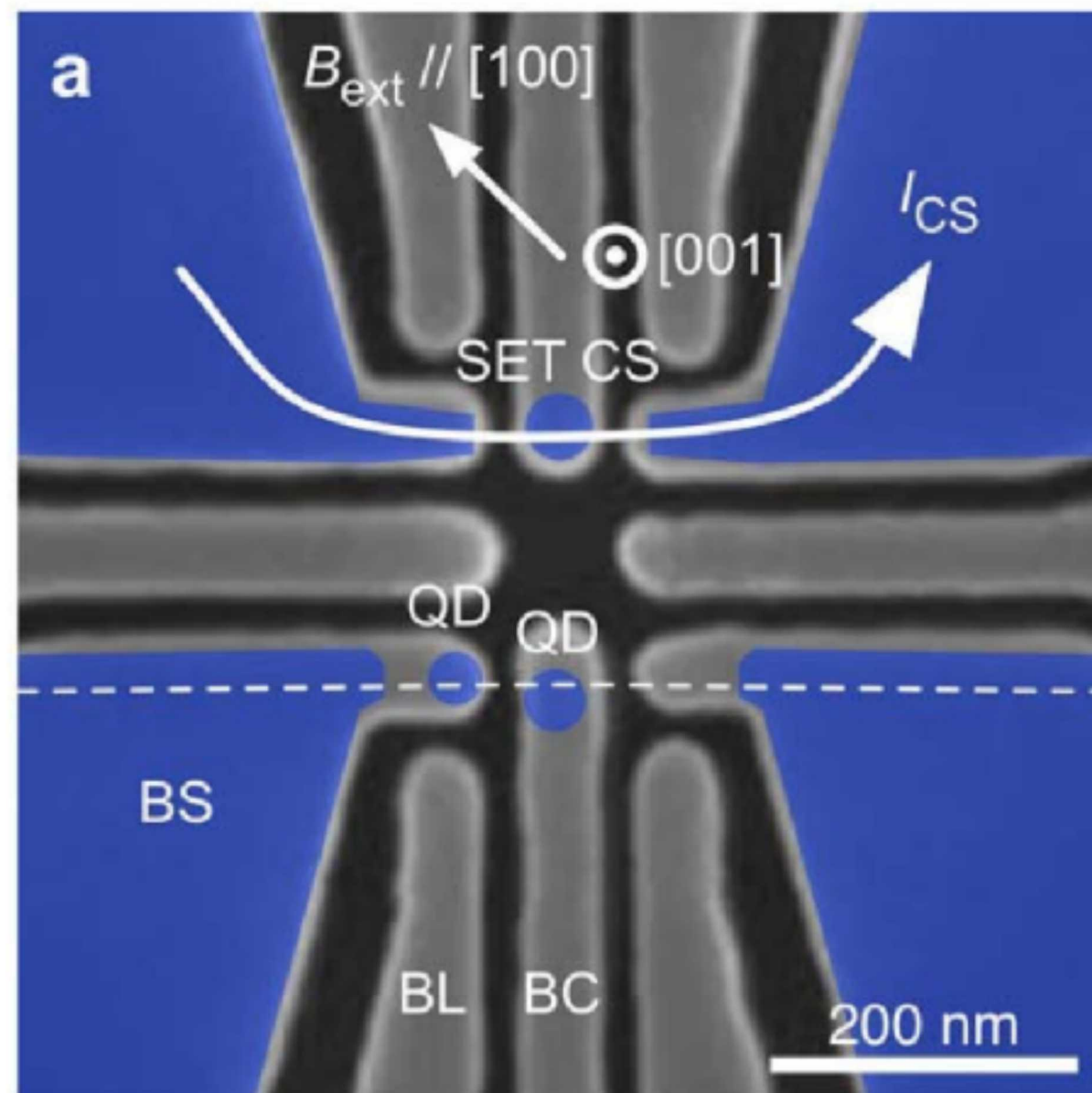
There are a number of interesting **open questions** we are just starting to work on

1. Does the unitary ansatz buy you any **additional accuracy**? If not, why use quantum?
2. How does noise affect the **truncation hierarchy**?
3. Assumes that I can efficiently prepare a reference state *and* apply the UCC circuit.
Classical reference state preparation is still NP-Complete, are we fooling ourselves?

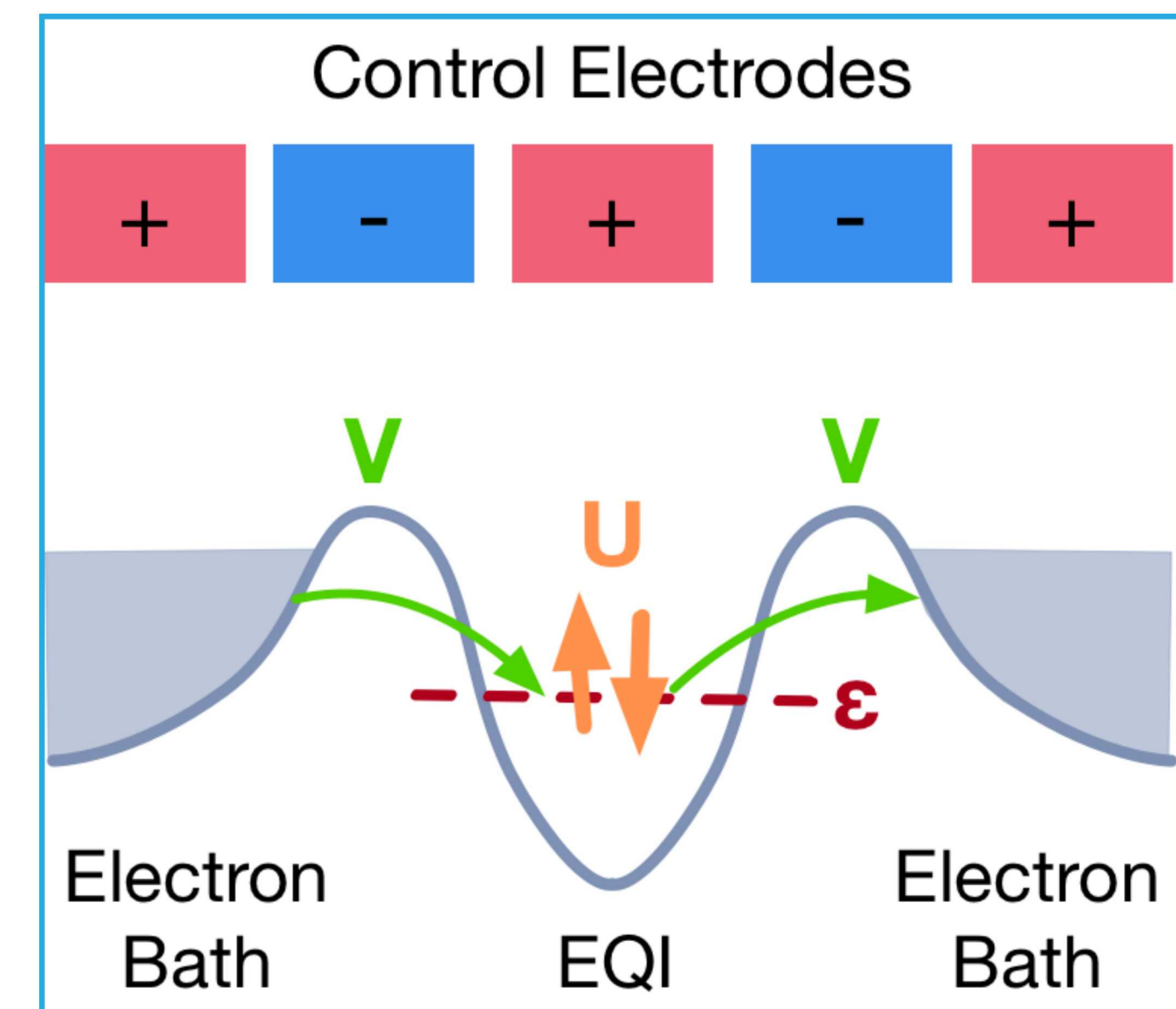
Quantum impurities in semiconductor devices

All-electrical universal control of a double quantum dot qubit in silicon MOS

Patrick Harvey-Collard,^{1,2} Ryan M. Jock,² N. Tobias Jacobson,² Andrew D. Baczewski,² Andrew M. Mounce,² Matthew J. Curry,^{2,3} Daniel R. Ward,² John M. Anderson,² Ronald P. Manginell,² Joel R. Wendt,² Martin Rudolph,² Tammy Pluym,² Michael P. Lilly,² Michel Pioro-Ladrière¹ and Malcolm S. Carroll²



Charge sensor: physical manifestation of a quantum impurity model



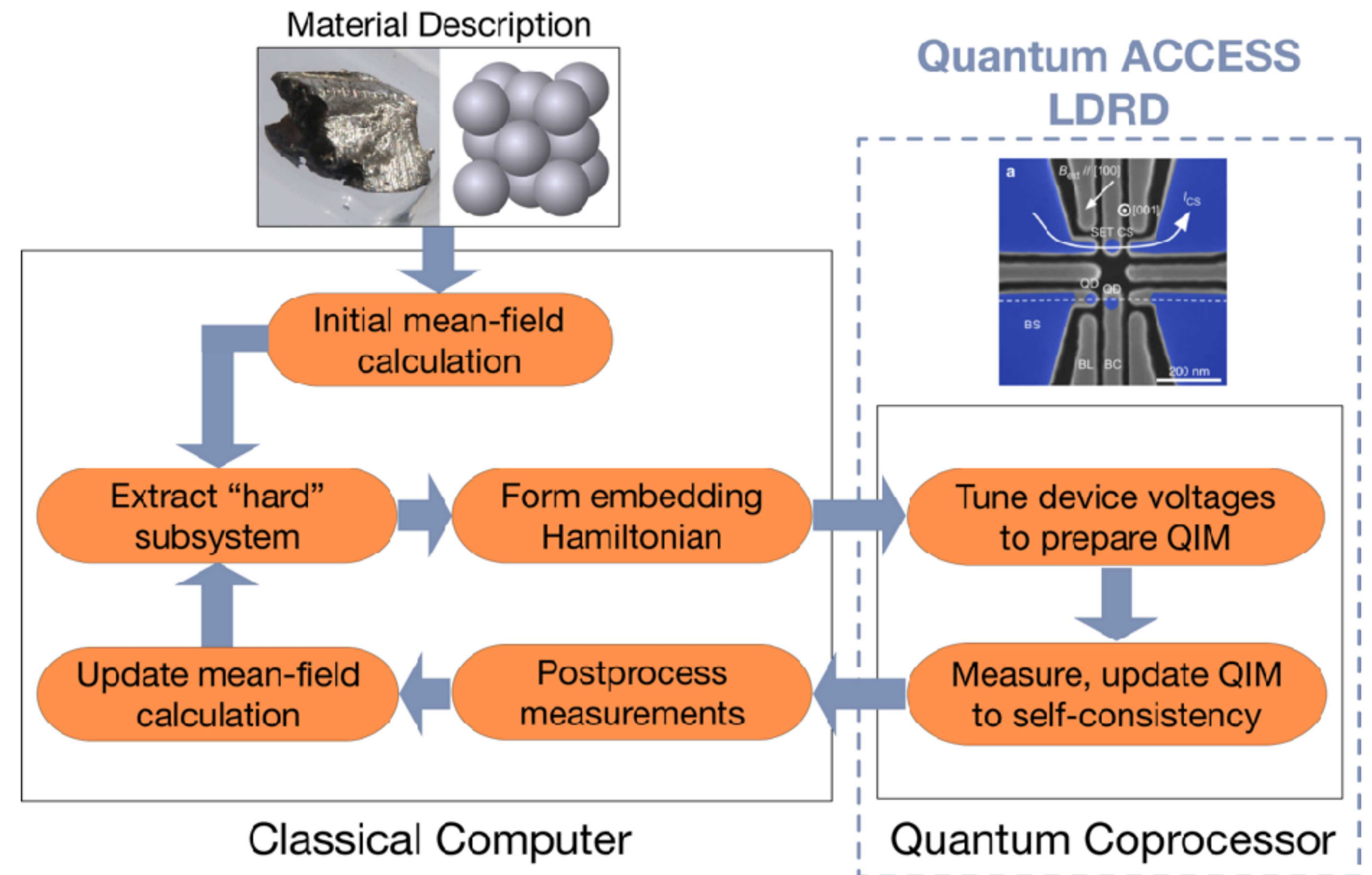
In the **strong coupling regime**, the **charge sensor** is more difficult to simulate than the **qubit**!

$$\hat{H}_{QIM} = \sum_{\sigma} \varepsilon_{\sigma} \hat{c}_{\sigma}^{\dagger} \hat{c}_{\sigma} + U \hat{c}_{\uparrow}^{\dagger} \hat{c}_{\uparrow} \hat{c}_{\downarrow}^{\dagger} \hat{c}_{\downarrow} + \sum_{\mathbf{k}, \sigma} \left(E_{\sigma}(\mathbf{k}) \hat{b}_{\sigma}^{\dagger}(\mathbf{k}) \hat{b}_{\sigma}(\mathbf{k}) + V_{\sigma}(\mathbf{k}) \left(\hat{c}_{\sigma}^{\dagger} \hat{b}_{\sigma}(\mathbf{k}) + \text{h.c.} \right) \right)$$

Hybrid analog dynamical mean-field theory (DMFT)

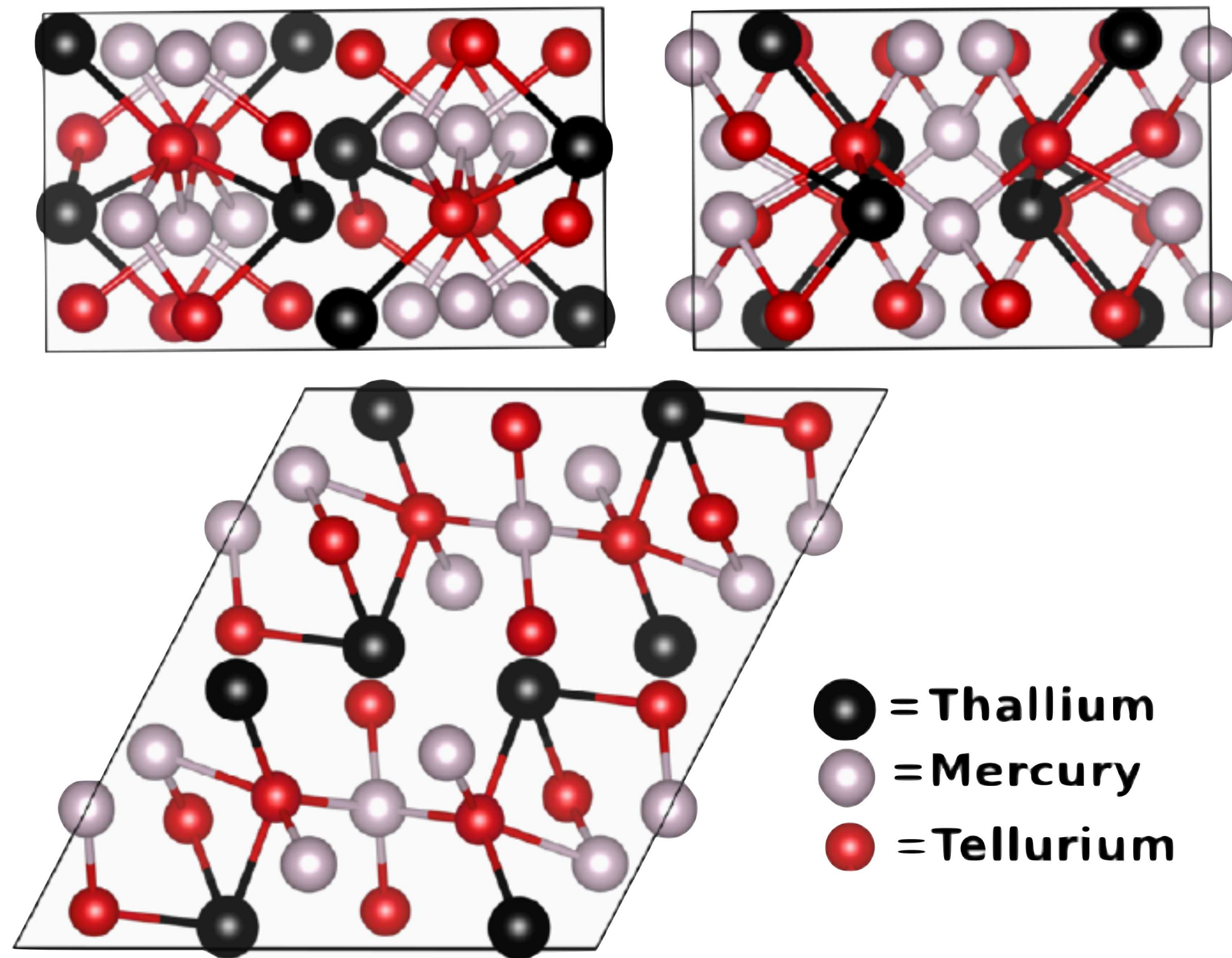
DMFT iterates between an
“easy” mean-field problem and a
“hard” quantum impurity problem

Can we use measurements of a
strongly coupled quantum impurity
to “solve” the quantum impurity
problem?

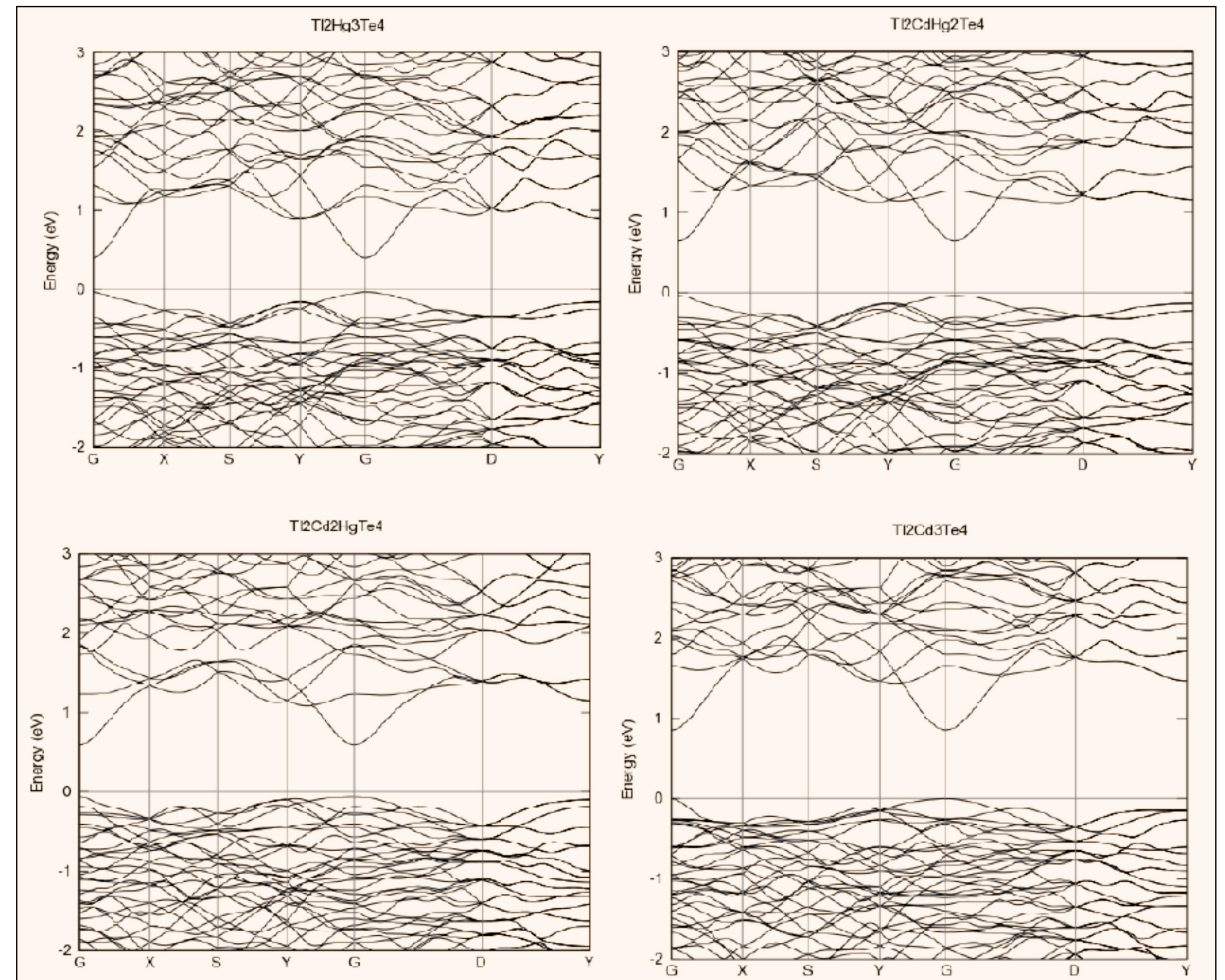


You’ll hear a bit more about this during [Mitchell Brickson’s update talk \(December 5\)](#)

An anecdote in closing



Unit cell of $Tl_2Hg_3Te_4$



“The computer doesn’t do physics. You do.”

Conclusion

- Quantum simulation has a promising future, but there are many lessons to be learned from 70 years of computational chemistry / materials science
- We've discussed:
 - How **DFT** helps us design semiconductor-based qubits
 - Comparisons of **QMC** and **DFT** that indicate robustness of certain properties
 - Instances of **TDDFT** being used to constrain experiments and how NISQ HW might help
 - How higher levels of theory (e.g., **CCSD**) help us justify using less expensive ones
 - A hybrid approach to **DMFT** that amplifies the power of analog simulation

Thank you for your time!

The views expressed in this presentation do not necessarily represent the views of the U.S. Department of Energy or the United States Government.



Sandia National Laboratories is a multi-missions laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for DOE's National Nuclear Security Administration under contract DE-NA0003525.

