Research Title: Ab Initio Modeling of Point Defects for Quantum Information Science

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Academic Area/Field and Education Level
Physics, Chemistry, Materials Science Engineering, or equivalent (MS or PhD level)

Objectives: Develop quantum chemical models of novel point defects in wide bandgap semiconductors. Predict absorption energy, emission wavelength, excited state lifetime, spin and hyperfine properties, and the dependence of these quantities on electric, magnetic, and strain fields.

Description: Our group is interested in novel point defects in wide bandgap semiconductors which can serve as qubit systems for quantum sensing. The implementation of quantum sensors in solid state systems will create new possibilities for integration of these capabilities in Air Force platforms. We are specifically interested in systems that show optical activity under the influence of electric, magnetic, and strain fields. A useful starting point for thinking about the problem is the work by Weber *et al.*, which outlines a general strategy for looking for material systems that could have optically detected magnetic resonance (ODMR) [1]. In general, defects in tetrahedral coordinated semiconductors have symmetry properties which are promising for ODMR. This work motivated explorations of defects beyond the nitrogen-vacancy (NV) center in diamond to defects in SiC ($V_{Si}V_C$, V_{Si}, and Cr_{Si} [2,3]) and the germanium-vacancy center in diamond [4]. There have been fewer reports of quantum sensing in these newer systems; however, quantum sensing requires full understanding of the spin properties and of hyperfine interactions in such systems. Experiment can deduce only certain system characteristics, and it is laborious.

Complementary to experimental characterization of hyperfine interactions, *ab initio* quantum modeling has also shown to be effective in calculating the characteristics of hyperfine interactions of NV centers with surrounding nuclear spins [5]. Supercell techniques treat long-range defect wave-function tails properly by employing periodic boundary conditions, but each cell replicates the defect, introducing interaction artifacts. In contrast, supercluster techniques model a single non-interacting defect, but the cluster must be of sufficient size to avoid constraining the bounds of the wave function artificially. Hyperfine interactions in diamond NV centers have been modeled using several common techniques, with the most reliable studies using either: (1) a 512-atom *supercell* treatment [6,7], or, alternatively (2) $C_{84}[NV]H_{78}$ [8], or $C_{291}[NV]H_{172}$ [9] hydrogen-terminated *supercluster* approaches. This is an unimaginable task

for experimentalists. A very recent $C_{510}[NV]H_{252}$ study has shown good comparison with measurements of NV spin flips induced by neighboring ^{13}C nuclear spins [10].

Recently ab initio theory has proven essential to the proper characterization of electronic structures and observable properties of interesting defect centers. For the SiC divacancy, new predictions about the defect configuration [11], combined with the proposal of a new mechanism of the SiC divacancy photoluminescence [12], reinforces the need for treatment employing highly accurate *ab initio* methods. Zeeman splitting of the silicon vacancy in SiC was successfully modeled by Hepp *et al.* [13] using a group theoretical approach. Meanwhile other groups are using DFT to compute EPR parameters [14]. Thus the computational techniques are already established and the race is on to predict and fabricate designer defects with highly refined properties.

The magnetic properties of many other defects are not known. Identifying and designing candidate physical systems for use in quantum sensing are critical steps in the development cycle. High-throughput methods for screening optical and spin properties are needed. The problem is that the current state-of-the-art in modeling defect spectroscopy is mostly reliant on density functional theory (DFT), which can achieve accuracies of 0.2–0.3 eV for optical transitions. Much higher accuracy is needed to be useful in predicting the spin-dependent optical transitions of point defects. This project will develop cluster, supercell and hybrid quantum-mechanical/molecular-mechanical methods for efficient screening of new defect centers in e.g., diamond, SiC, GaN, AlN, and □-Ga2O3. The goal is to calculate absorption energy, emission wavelength, excited state lifetime, spin and hyperfine properties of these defects, as well as dependence of these quantities on electric, magnetic, and strain fields. The results of these calculations will be used to guide ongoing experimental work to synthesize and characterize these centers.

Research Classification/Restrictions: Unclassified, unrestricted.

Eligible Research Institutions: All U.S. institutions with graduation programs in the relevant academic areas listed above. Coordination with the topic sponsor is encouraged prior to proposal submission.

References:

[1] J. R. Weber, W. F. Koehl, J. B. Varley, et al., "Quantum computing with defects," PNAS **107**, 8513–8518 (2010).

[2] W. F. Koehl, B. Diler, S. J. Whiteley, et al., "Resonant optical spectroscopy and coherent control of Cr4+ spin ensembles in SiC and GaN," Phys. Rev. B **95**, (2017).

[3] A. Lohrmann, B. C. Johnson, J. C. McCallum, et al., "A review on single photon sources in silicon carbide," Rep. Prog. Phys. **80**, 034502 (2017).

[4] T. Iwasaki, F. Ishibashi, Y. Miyamoto, et al., "Germanium-vacancy single color centers in diamond," Scientific Reports **5**, 12882 (2015).

[5] B. Smeltzer, L. Childress, and A. Gali, "^{13}C hyperfine interactions in the nitrogen-vacancy centre in diamond," New Journal of Physics **13**, 025021 (2011).

[6] A. Gali, M. Fyta, and E. Kaxiras, "Ab initio supercell calculations on nitrogen-vacancy center in diamond: electronic structure and hyperfine tensors," Phys. Rev. B **77**, 155206 (2008).
[7] A. Gali, "Identification of individual C13 isotopes of nitrogen-vacancy center in diamond by combining the polarization studies of nuclear spins and first-principles calculations," Phys. Rev. B **80**, 241204 (2009).
[8] A. P. Nizovtsev, S. Ya. Kilin, P. Neumann, et al., "Quantum registers based on single NV + n ^{13}C centers in diamond: ii. spin characteristics of registers and spectra of optically detected magnetic resonance," Optics and Spectroscopy **108**, 239–246 (2010).
[9] A. P. Nizovtsev, S. Ya Kilin, A. L. Pushkarchuk, et al., "Theoretical study of hyperfine interactions and optically detected magnetic resonance spectra by simulation of the C_{291} [NV] $^{-}$ H_{172} diamond cluster hosting nitrogen-vacancy center," New Journal of Physics **16**, 083014 (2014).
[10] A. P. Nizovtsev, S. Y. Kilin, A. L. Pushkarchuk, et al., "Non-flipping ^{13}C spins near an NV center in diamond: hyperfine and spatial characteristics by density functional theory simulation of the C_{510} [NV]H_{252} cluster," New Journal of Physics **20**, 023022 (2018).
[11] J. Davidsson, V. Ivády, R. Armiento, et al., "First principles predictions of magneto-optical data for semiconductor defects: the case of divacancy defects in 4H-SiC," New Journal of Physics **20**, 023035 (2018).
[12] B. Magnusson, N. T. Son, A. Csóré, et al., "Excitation properties of the divacancy in 4H-SiC," ArXiv:1804.01167 [Cond-Mat] (2018).
[13] C. Hepp, T. Müller, V. Waselowski, et al., "Electronic structure of the silicon vacancy color center in diamond," Phys. Rev. Lett. **112**, 036405 (2014).
[14] A. Komarovskikh, A. Dmitriev, V. Nadolinny, et al., "A DFT calculation of EPR parameters of a germanium-vacancy defect in diamond," Diamond and Related Materials **76**, 86–89 (2017).