Two-dimensional Magnetic Metal Organic Frameworks: Computational Study of Electron and Phonon Transport

A. Jalali, N. Vlastos, M. Mahmoudi Gahrouei, L. de Sousa Oliveira
Department of Chemistry, University of Wyoming, 1000 E. University Ave, Laramie Wyoming 82071
e-mail: ajalali@uwyo.edu

INTRODUCTION

Magnetic metal organic frameworks (MOFs) are excellent candidates for electronic and spintronic applications due to their tunable structures, high porosity, and, in some cases, strong electron correlation. MOFs provide structural flexibility through precise tuning of their electronic and magnetic properties by selecting specific ligands and metal nodes [1]. These materials exhibit diverse charge transport mechanisms such as band-like conduction facilitated by π-conjugated linkers and hopping transport mediated by metal centers [2, 3]. MOFs have an inherently low thermal conductivity due to their high porosity and weak bonding [4]. Suppressing heat conductivity while maintaining reasonable charge transport is advantageous for achieving a higher thermoelectric figure of merit (ZT). Understanding both electron and phonon transport in magnetic MOFs is essential for evaluating their performance in electronic and thermoelectric devices. This study aims to provide a benchmark study of electron and phonon transport behavior in transition metal-based 2D MOFs.

METHODOLOGY

We investigate the electronic and phononic transport properties of magnetic MOFs using self-consistent charge density functional tight binding (SCC-DFTB) and density functional theory (DFT). For electronic transport, we examine essential electronic properties such as band structure, density of states (DOS), electrical conductivity, and Seebeck coefficient in MOFs with transition metal nodes like Fe, Co, and Ni. Phonon transport calculations, including lattice thermal conductivity, are also obtained, allowing us to provide estimates for the thermoelectric figure of merit (ZT) of such MOFs. In addition, we begin exploring strategies such as doping to enhance charge mobility since MOFs exhibit low intrinsic electrical conductivity. Electron mobility is estimated via the Boltzmann transport equation within the constant relaxation time approximation.

CONCLUSION

By benchmarking SCC-DFTB against DFT, we aim to quantitatively evaluate the trade-off between computational efficiency and predictive accuracy in modeling both electron and phonon transport in magnetic 2D MOFs.

ACKNOWLEDGMENT

The authors acknowledge the computational resources provided by the University of Wyoming.

REFERENCES

[1] V. Rubio-Giménez, S. Tatay, and C. Martí-Gastaldo, Electrical conductivity and magnetic bistability in metal–organic frameworks and coordination polymers: charge transport and spin crossover at the nanoscale, Chemical Society Reviews **49**, 5601 (2020).

[2] T. Musho and N. Wu, Ab initio calculation of electronic charge mobility in metal–organic frameworks, Physical Chemistry Chemical Physics **17**, 26160 (2015).

[3] M. Kozlowska, Y. Pramudya, M. Jakoby, S. Heidrich, L. Pan, B. S. Richards, I. A. Howard, C. Wöll, R. Haldar, and W. Wenzel, Crystalline assembly of perylene in metal–organic framework thin film: J-aggregate or excimer? Insight into the electronic structure, Journal of Physics: Condensed Matter **33**, 034001 (2020).

[4] B. Huang, A. McGaughey, and M. Kaviany, Thermal conductivity of metal-organic framework 5 (MOF-5): Part I. Molecular dynamics simulations, International Journal of Heat and Mass Transfer **50**, 393 (2007).