O 70: Poster Session V: Poster to Mini-Symposium: Frontiers of electronic-structure theory II

Time: Wednesday 10:30–12:30 Location: P

O 70.1 Wed 10:30 P

The coordination corrected enthalpies method in AFLOW — •Rico Friedrich[1,2], Marco Esters[1], Corey Oses[1], Stuart Ki[1], Maxwell J. Brenner[1], David Hicks[1], Michael J. Mehl[1], Mahdi Ghorbani-Asl[2], Arkady Krasheninnikov[2], Cormac Toher[1], and Stefano Curtarolo[1,3] — [1]Center for Autonomous Materials Design, Duke University, USA — [2]Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany — [3]Materials Science, Electrical Engineering, Physics and Chemistry, Duke University, USA

The AFLOW database and software leverages *ab initio* calculations for autonomous materials design [1]. The predictive power critically relies on accurate formation enthalpies - quantifying the thermodynamic stability of a compound. For polar materials such as chalcogenides (*e.g.* oxides), pnictides (*e.g.* nitrides), and halides, standard semi-local DFT leads to errors of several hundred meV/atom [2,3].

We have recently developed the "coordination corrected enthalpies" (CCE) method yielding highly accurate room temperature formation enthalpies with mean absolute errors down to 27 meV/atom [3]. Here, we introduce AFLOW-CCE: a tool where users can input a structure file of their system and receive the CCE corrections, or even the CCE formation enthalpies if pre-calculated LDA, PBE or SCAN formation enthalpies are provided. The results can be used for the computational design of *e.g.* 2D materials.

[1] S. Curtarolo *et al.*, Comput. Mater. Sci. **58**, 218 (2012).
[2] V. Stevanović *et al.*, Phys. Rev. B **85**, 115104 (2012).
[3] R. Friedrich *et al.*, npj Comput. Mater. **5**, 59 (2019).

O 70.2 Wed 10:30 P

Non-perturbative modelling of exciton-phonon interactions in organic semiconductors — •Antonios M. Alvertis[1], Timothy J.H. Hele[2], and Bartomeu Monserrat[3] — [1]Cavendish Laboratory, University of Cambridge, J.,J.,Thomson Avenue, Cambridge CB3 0HE, United Kingdom — [2]Department of Chemistry, University College London, 20, Gordon Street, London WC1H 0AJ, United Kingdom — [3]Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge CB3 0FS, United Kingdom

The optoelectronic response of organic semiconductors is dominated by excitons, which often strongly interact with phonons. This makes perturbation theory insufficient for describing exciton-phonon interactions in these materials. Here we present a non-perturbative methodology for accurately capturing exciton-phonon interactions to all orders, combining finite differences methods for phonons with GW-BSE or TD-DFT calculations for crystalline or isolated systems respectively. We show that exciton spatial delocalisation is the main parameter controlling the magnitude of its coupling to phonons and reveal the microscopic mechanism of exciton temperature- and pressure-dependence in molecular crystals. Furthermore, accounting for nuclear quantum motion is found to be critical for achieving quantitative accuracy in the prediction of exciton energies, both for single molecules and periodic systems. Overall, we provide a unified picture of exciton-vibration interactions in organic semiconductors, reconciling the complementary views of finite molecular clusters and periodic molecular solids.

O 70.3 Wed 10:30 P

Ultrafast control of material optical properties via the infrared-resonant Raman effect — •Guru Khalsa, Nicole Benedek, and Jeffrey Moses — Cornell University, Ithaca, NY, USA

The Raman effect - the inelastic scattering of light by lattice vibrations - provides an important tool for conversion of light from one color to another in optical physics, and is ubiquitous in materials characterization because of its fundamental connection to crystal symmetry. The Raman effect is dominated by changes to the electronic susceptibility in the UV and visible frequency ranges. However, in the mid- and far-IR, nonlinear contributions to the lattice polarization provide additional Raman pathways, which have been little explored.

Using a combination of theory and first-principles calculations, we derive and evaluate symmetry relations and complete expressions for the Raman effect in insulating crystalline materials including all electronic and lattice-mediated pathways. We show that when infrared-active phonons are resonantly excited, the Raman effect may be dominated by direct changes to the lattice polarizability induced by Raman phonons. Applying this approach to the archetypal perovskite SrTiO3, we show that this infrared-resonant Raman effect can induce optical symmetry breaking and giant refractive index shifts that are tailored by the incident light polarization and which infrared active phonons are excited. (arXiv:2011.02010)

O 70.4 Wed 10:30 P

Non-Perturbative Theory of Charge Transport in Crystalline Solids — •Christian Carbogno and Matthias Scheffler — Fritz-Haber-Institut der Max-Planck-Gesellschaft

Our understanding of charge transport in crystalline solids predominantly relies on the Boltzmann transport equation. However, its perturbative approximations for the nuclear dynamics and for its coupling to the electrons can be inaccurate in complex materials [1]. We present an alternative, non-perturbative *ab initio* Green-Kubo approach based on a new formulation of the flux viz. polarization. It can be evaluated via *ab initio* molecular dynamics and only requires gauge-fixed properties. At variance with Berry phase approaches [2], it can thus be evaluated for (semi-)conductors at finite temperatures featuring thermal electronic excitations. We demonstrate our methodology by calculating the electrical conductivity for 2D honeycomb lattices as well as for the harmonic material Si and the anharmonic perovskite SrTiO$_3$. Furthermore, we systematically compare to non-perturbative Kubo-Greenwood calculations and discuss why the latter approach has so far only been numerically applicable for materials with strong structural disorder [3], i.e., when the dispersion in reciprocal space is negligible.

[1] M. Zacharias, M. Scheffler, and C. Carbogno, *Phys. Rev. B* **102**, 045126 (2020).
[2] R. D. King-Smith and D. Vanderbilt, *Phys. Rev. B* **47**, 1651 (1993).
[3] B. Holst, M. French, and R. Redmer, *Phys. Rev. B* **83**, 235120 (2011); C. Di Paola, *et al.*, *Phys. Rev. Research* **2**, 033055 (2020).

O 70.5 Wed 10:30 P

Uncovering the Relationship Between Thermal Conductivity and Anharmonicity with Symbolic Regression — •Thomas Purcell, Matthias Scheffler, Luca Ghiringhelli, and Christian Carbogno — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Quantitatively understanding the link between anharmonicity and thermal conductivity, κ, is pivotal to the search for better thermal insulators. While it is qualitatively known that more anharmonic materials have a lower κ, until recently, no quantitative measure of anharmonicity existed. Here we present descriptors of κ based on our new measure of anharmonicity, σ^{A} [1]. We find the analytical expressions with symbolic regression, via the sure-independence screening and sparsifying operator (SISSO) method [2]. To better capture the nonlinearities in the correlation between κ and σ^{A}, we introduce an automatic scaling and shifting of the input data when generating new features like $\exp(\alpha x + a)$. Using our new strategy, we generate expressions that are competitive with those previously reported in the literature using only a third of primary the features [3], and reduce the test error of the models when compared to traditional SISSO. Finally, we discuss the implications of the new models on future materials design.

[1] F. Knoop, et al. Phys. Rev. Mat. 4. 083809 (2020)
[2] R. Ouyang, et al. Phys. Rev. Mat. 2. 083802 (2018)
[3] L. Chen, et al., J. Com. Mat. Si. 170. 109155 (2019)

O 70.6 Wed 10:30 P

Excited Electron Enhanced Defect Diffusion in MgO under Ultrafast Laser Irradiation — •Yifan Yao and Andre Schleife — University of Illinois, Urbana-Champaign

Electronic excitation of defect states has been predicted to enhance ion diffusion via manipulating the explicit electron distribution. Due to the induced non-linear excitation, the ultrafast laser can offer a novel opportunity to control the electron dynamics in materials. Hence, a detailed understanding of laser-materials interaction is vital for manufacturing with nanoscale precision but still remains unclear, due to its non-linear and non-equilibrium character. Here, we use oxygen vacancy migration in MgO as an example to study the ion diffusion in laser-irradiated materials. We apply real-time TDDFT, which can accurately describe such nonlinear effects, to calculate the time-evolution of the occupation number under different laser frequencies and intensities. Comparison to the distribution of hot electrons following proton irradiation can provide insights into how diffusion enhancement can be achieved by the transient localized electron dynamics and how it depends on specific laser parameters.

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O 70.7 Wed 10:30 P

Modelling the structural and charge transport properties of merocyanine single-crystals

— •Nora Gildemeister[1], Fabrizia Negri[2], Klaus Meerholz[1], and Daniele Fazzi[1] — [1]Insitut für Physikalische Chemie, Department Chemie, Universität zu Köln, Greinstr. 4-6, D - 50939 Köln — [2]Dipartimento di Chimica, Università di Bologna, via F. Selmi 2, 40126 Bologna, Italy

Merocyanines are highly polar organic π-conjugated molecules investigated for their self-assembly and optoelectronic properties. The accurate description of their molecular, electronic and vibrational structure remains a challenge due to strong electron correlation effects and long-range inter-molecular interactions. [1-2]

We report a comprehensive analysis modelling intra- and inter-molecular charge transport properties for a library of different donor-acceptor units and lateral groups. We found that constrained DFT is an effective embedding method to correctly assess the molecular and electronic structure in single crystals. Charge mobilities were computed within the semiclassical nonadiabatic electron-transfer theory by analysing different single crystals and highlighting the impact of side groups and casting conditions. Computed and experimental values are in good agreement. Our modelling suggests that charge diffusion is maximized when dipolar molecules are packed in slipped anti-symmetric pairs, arranged in 2D interconnected architectures. [3]

[1] C. Brückner, et al., J. Phys. Chem. C 2015, 17602-17611. [2] D. Bialas, et al., J. Phys. Chem. C 2019, 123, 30, 18654-18664. [3] N. Gildemeister, et al. Paper in preperation.