

O 108: Poster Session VIII: Poster to Mini-Symposium: Frontiers of electronic-structure theory

V

Time: Thursday 13:30–15:30

Location: P

O 108.1 Thu 13:30 P

Electron-phonon band gap renormalization in graphene nanoribbons — ●RODRIGO MENCHÓN¹ and ARAN GARCIA-LEKUE^{1,2} — ¹Donostia International Physics Center (DIPC), San Sebastian, Spain — ²Ikerbasque, Basque Foundation for Science, Bilbao, Spain

During the last decade, on-surface synthesis techniques have paved the way to the creation of atomically precise carbon-based nanostructures, e.g. 1D stripes referred to as graphene nanoribbons (GNRs).[1] Alongside these experimental advancements, first-principle simulations have been pivotal for understanding and predicting the electronic properties of GNRs. In particular, GNRs with so-called armchair edges are specially well-studied due to their semiconductor character.[2,3] This makes them very promising for electronic applications, and, at the same time, demands a high quality description of their semiconducting gap. Within this scenario, it would be interesting to address electron-phonon induced energy renormalization effects and their dependence upon temperature.[4] In this work, starting from Density Functional Theory simulations the electron self-energies and the band gap renormalization of armchair GNRs resulting from the electron-phonon interactions are determined. Our method might be extended to other semiconducting families of GNRs, 0D systems (e.g. carbon macromolecules) or 2D materials (e.g. nanoporous graphene).

References: [1] S. Clair et al., Chem. Rev. 2019, 119, 7, 4717 (2019). [2] L. Talirz et al., Adv. Mater. 2016, 28, 6222 (2016). [3] J. Lawrence et al., ACS Nano 2020, 14, 4, 4499 (2020). [4] F. Brown-Altwater et al., Phys. Rev. B 101, 165102 (2020).

O 108.2 Thu 13:30 P

Anomalous low-velocity electronic stopping in proton-irradiated graphene — ●ALINA KONONOV¹ and ANDRE SCHLEIFE² — ¹Department of Physics, University of Illinois at Urbana-Champaign — ²Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign

Ion beams in high-resolution imaging and patterning techniques can be used efficiently to manipulate and characterize 2D materials. Low particle velocities are particularly interesting, since in this regime the drag force induced on an incident ion by electrons is rife with material-specific effects. Examples include the threshold velocity for semiconductors and insulators below which an ion cannot excite electrons across the band gap and electronic stopping vanishes. In addition, directional bonding in these materials makes electronic stopping sensitive to the ion's trajectory even for slow ions, when core electrons are negligible. Graphene, thus, presents a highly interesting case with directional bonding but no band gap. Our real-time time-dependent density functional theory simulations of proton-irradiated graphene reveal a shoulder in the low-velocity stopping of channeling protons which does not occur for protons traversing a centroid path. From analyzing the post-impact band occupations and projectile charge state, we infer that resonant charge capture from certain valence bands by channeling protons is responsible for this feature. Our prediction of a new form of anomalous low-velocity stopping has implications for ion beam imaging, where such trajectory-dependent behavior could be exploited to achieve high resolution. (Supported by NSF OAC 17-40219)

O 108.3 Thu 13:30 P

Ultrafast dynamics of hot carriers in bulk semiconductors and in accumulation layer: energy relaxation and screening effects. — ●JELENA SJAKSTE — LSI, CEA/DRF/IRAMIS, CNRS, Ecole polytechnique, Institut Polytechnique de Paris, 91120 Palaiseau, France

Electron-phonon coupling determines the charge transport properties in pure materials as well as the relaxation dynamics of photoexcited carriers. The rapid development of the computational methods based on density functional theory, on the one hand, and of the time- energy- and momentum- resolved spectroscopy, on the other hand, allows today an unprecedentedly detailed insight into the role of the electron-phonon coupling [1,2].

In this work, we will present our recent results, both experimental and theoretical, on hot electron relaxation in silicon [3]. Moreover, we will present our recent results, both experimental and theoretical, on the hot electron relaxation in InSe. InSe is a quasi-2D material which

was shown recently to have potential interest for optoelectronics. In this work, we will discuss our new results on the relaxation dynamics and screening of the electron-phonon interaction in doped InSe [4].

References:

- [1] J. Sjakste et al, J. Phys: Cond. Mat. 30, 353001 (2018).
- [2] Tanimura et al, Phys. Rev. B 93, 161203 (R) (2016).
- [3] Tanimura, Kanasaki, Tanimura, Sjakste, Vast, Phys. Rev. B 100, 035201 (2019).
- [4] Chen, Sjakste et al, PNAS 117, 21962-21967 (2020).

O 108.4 Thu 13:30 P

Impact of the structural dynamics on the electronic properties of the metal organic frameworks — ●MARIANA KOZŁOWSKA¹, RITESH HALDAR², SHAHRIAR HEIDRICH¹, MARCUS ELSTNER³, CHRISTOF WÖLL², and WOLFGANG WENZEL¹ — ¹Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), Germany — ²Institute of Functional Interfaces (IFG), KIT — ³Institute of Physical Chemistry (IPC), KIT

Electronic properties of organic semiconductors (OSC) depend on the assembly of molecules and their vibrational flexibility in a material. These determine the microscopic intercommunication, resulting in a materials conduction. Here, we have employed a metal-organic framework (MOF) type of assembly strategy to engineer the arrangement of the (i) OSC pentacene and (ii) the DPA-TPE (diphenylamine-tetraphenylethylene) chromophore, and demonstrated the change of the electronic and spectroscopic properties of molecules and films towards assembly in the spatially ordered MOF structure.

Using the combination of DFT, DFT-B, QM/MM methods, the charge carrier mobility, starting from the band structure and the Marcus charge hopping, to the direct propagation of charge carriers using the time dependent Schrödinger equation, coupled to the classical motion of nuclei, were performed. This allowed to identify localized frustrated rotations of the pentacene, as the reason for the breakdown of band transport, and constraining of rotations in DPA-TPE, leading to the TADF emission upon crystallization in MOF.

O 108.5 Thu 13:30 P

Tailoring the electronic anisotropy of nanoporous graphene — ●XABIER D. DE CERIO¹ and ARAN GARCIA-LEKUE^{1,2} — ¹Donostia International Physics Center (DIPC), San Sebastian, Spain — ²Ikerbasque, Basque Foundation for Science, Bilbao, Spain

Recent experimental advances have allowed the synthesis of atomically precise nanoporous graphene (NPG) through the lateral fusion of graphene nanoribbons (GNRs). Besides being a semiconductor, this novel 2D material exhibits a highly anisotropic electronic structure, which makes it appealing for electronic, optical, and sensing applications, and has thus generated great interest [Science 360, 199 (2018); Nano Lett. 19, 576 (2019); J. Am. Chem. Soc. 141, 13081 (2019)]. In this work, by means of Density Functional Theory (DFT) calculations, we determine the electronic anisotropy of a new family of NPG, in which atomic scale modification of lateral linkers leads to three different inter-ribbon coupling configurations. Our calculations predict different anisotropic behaviour for each coupling configuration. Additionally, we find that the rotation angle of phenyl rings in the linkers works as a continuous knob to manipulate the electronic crosstalk between adjacent GNRs. Supported by on-surface synthesis of this new nanostructure, we present a proof-of-concept study showing that engineering the linkers is an efficient way to tune the electronic anisotropy of NPG.

O 108.6 Thu 13:30 P

Topological semimetallic phase in PbO₂ promoted by temperature — ●BO PENG¹, IVONA BRAVIC¹, JUDITH MACMANUS-DRISCOLL², and BARTOMEU MONSERRAT^{1,2} — ¹Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom — ²Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge CB3 0FS, United Kingdom

The past decade has witnessed multiple experimental realization and thousands of predictions of topological materials. However, it has been determined that increasing temperature destroys topological order, re-

stricting many topological materials to very low temperatures and thus hampering practical applications. Here, we propose the first material realization of temperature promoted topological order. We show that a semiconducting oxide that has been widely used in lead-acid batteries, β -PbO₂, hosts a topological semimetallic phase driven by both thermal expansion and electron-phonon coupling upon increasing temperature. We identify the interplay between the quasi-two-dimensional

nature of the charge distribution of the valence band with the three-dimensional nature of the charge distribution of the conduction band as the microscopic mechanism driving this unconventional temperature dependence. Thus, we propose a general principle to search for and design novel topological materials whose topological order is stabilized by increasing temperature. This provides a clear roadmap for taking topological materials from the laboratory to technological devices.