Mini-symposium B4  
Molecular Scale Electronics/Spintronics

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B04-01 Keynote
Quantum mechanical simulation of transient currents through molecular devices
Guanhua Chen  
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In an open electronic system, the number of electrons is not conserved and often fractional, and thus, traditional quantum chemistry methods are no longer applicable. Based on the holographic electronic density theorem, we proposed and developed a first-principles method to simulate the dynamics of open electronic systems. A Liouville-von Neumann equation for reduced single-electron density matrix, coupled with Poisson equation, is numerically integrated in the time domain to simulate electronic dynamics of open systems. Transient currents through molecular transistor, carbon nanotube, graphene and molecular wires are simulated and studied.

B04-02 Invited
Electronic, Magnetic and Transport Properties of MoS2 and its Hybrid Structures
Zilu Wang, Qian Chen and Jinlan Wang  
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As a typical transition-metal dichalcogenide material, MoS2 has attracted intense research interest due to its promising electronic and optical properties. In this talk, I will present our recent results on thin MoS2 nanosheets by using density functional theory calculations: 1) defect-induced hopping transport and photoluminescence (PL) enhancement; 2) electronic structure of twisted bilayer of graphene and MoS2 and MoS2/MoS2; and 3) Pattern magnetic Co-doping on MoS2 nanosheet. Our calculations revealed that: 1) charge transport of few-layer MoS2 in low carrier density regime can be explained by hopping through defect-induced localized states and; ii) strong PL enhancement of monolayer MoS2 through defect engineering and oxygen bonding; iii) The twist of interlayers leads to significant variations in the band structure for both Gr/MoS2 and MoS2/Gr/MoS2 and the former shows clearly dependence on the rotation angle which is indeed from the twisted resultant lattice mismatch; iv) we predict a new 2D half-metallic Co/MoS2 heterojunction with robust ferromagnetic ground spin state and 100% spin current polarization and it is synthesizable via wetting deposition of Co on MoS2 by electron-beam evaporation technique.


B04-03 Invited
Ante Bilic  
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Electronic transport properties of several sets of narrow graphene nanoribbons will be presented, based on computational modeling. Charge transport through a two terminal device has been modeled using density functional theory and the non-equilibrium Green's function method. The predicted length dependence of the nanoribbons shows a very complex nature. Based on a low-resistance derivative of pyrene, two series of nanoribbons are generated, one demonstrating an exponential attenuation of conductance with length and the other set exhibiting a length and bias independent conductance. For an armchair series, based on the perylene motif, a similar length independent transport is predicted if thiols are used as the terminal groups. However, replacement of these by amino motifs yields an anomalous conductance, which increases with length. This unusual behavior is a consequence of the charge flow through the antisymmetric frontier electronic states with linear ("massless") dispersion, which can be effectively harnessed by multiple terminal groups. A similar anomalous conductance, which increases with length, but owing to a different mechanism, is also predicted for nanoribbons with zigzag edges. Finally, the potential of graphene meshes for gas sensing applications will be demonstrated. The gas molecules of NH3 and NO2 adsorb strongly on graphene, resulting in a sizeable electrical conductivity change.

B04-04 Invited
Effect of electron-vibron coupling on electron transport via single-molecule magnet Fe4  
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Recent experimental advances allow individual molecules to be placed between electrodes, and their electron transport to be measured at single-molecule junctions or transistors. One interesting family of molecules among them are anisotropic magnetic molecules referred to as single-molecule magnets (SMMs). A SMM comprises a few transition metal ions surrounded by several hundreds of atoms, with significant magnetic anisotropy. There have been studies of an interplay between quantum properties of SMMs and electron transport via individual SMMs. However, so far, the coupling between electric charge and vibrations of an SMM has not been theoretically examined in the context of single-molecule transistors. Here we investigate an effect of such coupling on electron transport via a SMM Fe4. We apply both density-functional theory (DFT) and model Hamiltonian approach, in the sequential tunneling regime. We show our calculated electron-vibron coupling strengths obtained from the DFT calculation and compare with experimental data [Burzuri et al., Nano Lett. 14, 3191 (2014)]. Additionally, we present current-voltage characteristics and differential conductance as a function of gate voltage, temperature, and applied magnetic field obtained from solving the master equation using the DFT-calculated parameter values.

B04-05 Invited
Electric and Thermoelectric Properties of Graphene Nanoribbons and Like Materials
Yao-Jun Dong, Ming-Xing Zhai, Xue-Feng Wang  
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The edge states of zigzag graphene nanoribbons (ZGNRs) and like materials have energies around the Fermi energy and are spontaneously spin polarized due to Coulomb interaction. The materials are then locally magnetized on edges and the antiferromagnetic (AFM) state with opposite edge magnetizations usually has a lower energy than the ferromagnetic (FM) state with parallel edge magnetizations. Because of the geometry symmetry in even-width nanoribbons, the materials show strong parity dependence of width. Chemical decorations or external fields can manipulate their physical properties in a wide variety. Employing density functional theory combined with the nonequilibrium Green's function method, we have studied the effects of doping and vacancies on spin transport
in ZGNRs, zigzag silicene and alpha-graphyne nanoribbons. We have confirmed or predicted that (1) Colossal magnetoresistance shows usually; (2) Giant magnetoresistance exists in even-width pristine or odd-width doped FM materials; (3) Single spin negative resistance appears in out-of-plane AFM materials doped by II- and III-group atoms; (4) Doping and vacancies can greatly enhance the spin Seebeck effect and thermal pure spin current may be realized; (5) Perfect current or spin diodes can be fabricated as well as perfect spin filters with the help of doping and geometry design.

**B04-06 Invited**

First-principles calculations of the magnetic anisotropic constants of Co–Pd multilayers: Effect of stacking faults

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Using first-principles density functional theory calculations with spin-orbit coupling, we systematically investigate the magnetic anisotropic energy (MAE) of CoₙPdₙ (n + m = 5) magnetic multilayers. We consider the influences of the relative atomic weight of Co, w_Co, stacking fault, and external stress on the MAE. We find that out-of-plane lattice constant, saturation magnetization, and magnetic moments are almost linearly correlated with w_Co. The magnetic anisotropic constant (MAC) curve of CoₙPdₙ without stacking fault shows a nearlinear dependence on w_Co that agrees with our derived effective MAC K_eff which includes shape, magneto-crystalline, and magneto-elastic contributions. We also show that the contributions from Pd layers to both the total magnetic moments and magnetic anisotropy are significant. The stress anisotropy due to the substrate has a weak effect on the MAC. However the stacking fault has a strong effect on the MAC. When the Co layer is thin, a Co–Pd interface without stacking fault is necessary for higher K_eff. However, when the Co layer is thick, creating stacking faults inside the Co region may produce a larger K_eff. Our study suggests the ways to increase the perpendicular magnetic anisotropy in Co–Pd multilayer systems and subsequently leads to the development of novel magnetic recording devices.

**B04-07 Keynote**

Rational Design of Functional Molecular Devices

R.Q. Zhang

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The continuous trend towards miniaturization of electronic devices makes organic molecular devices considered the most promising alternatives to conventional semiconductor devices. This is primarily because the transport properties of molecular junctions can be fine-tuned by varying their structures, facilitating the design of molecular devices with the necessary functionality. In this presentation, I describe our findings on the novel properties induced by hybrid state, intramolecular twist, side group, and molecular length in aromatic molecular junctions, based on calculations performed using the fully self-consistent nonequilibrium Green’s function method combined with density functional theory. Our work reveals the underlying mechanisms of these devices’ novel properties to be dependent on these modulating factors, underlining the high potential of rational design of functional molecular devices such as molecular rectifiers and switches.

**B04-08 Invited**

Band gaps and electric field effect in black phosphorus nanoribbons

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We study the energy band gaps of monolayer black phosphorus (phosphorene) nanoribbons (PNRs) and bandgap modulation by transverse electric fields via first-principles density functional theory. In contrast to graphene and MoS₂ nanoribbons, we find that 1) all hydrogen saturated PNRs, regardless of armchair or zigzag edges, are direct bandgap semiconductors, i.e., non-chirality; 2) the band gaps of armchair PNRs decrease monotonously without oscillation because of no symmetry-dependent edge states. The bandgaps of PNRs can be strongly modulated by transverse electric fields through a giant Stark effect, inducing a metal-insulator-transition, while they are not sensitive to perpendicular fields. Because of the giant Stark effect, a dual-gate PNR field-effect transistor can have high on/off ratio up to 10⁴ according to our transport calculations.

**B04-09 Invited**

Unusual electrical conductivity of materials with dual-nanoparticle inclusions

Jin-Cheng Zheng

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In this work, we study carrier scattering mechanisms and electronic transport properties of materials with dual-nanoparticle inclusions. Relaxation times for nanojunctions are obtained by calculating the transport cross section. Theoretical calculations are compared with experimental results for illustration of validation of computation. The origin of unusual temperature dependence of electrical conductivity, first-rise-then-fall, other than decreasing monotonically with increasing temperature in co-nanostructured materials is discussed.

**B04-10**

Transport through self-assembled-monolayer based molecular junctions

Yonatan Dubi

Department of Chemistry and The Ilze-Katz Institute for Nano-Scale Science and Technology, Ben-Gurion University of the Negev, Israel.

Self-assembled-monolayer (SAM) molecular junctions (MJs) constitute a promising building block candidate for future molecular electronic devices. Transport properties of SAM-MJs are usually calculated using either the phenomenological Simmons model, or a fully-coherent transport theory, employing the SAMs periodicity. As I will show, the standard theory seems to have some discrepancy with experimental observations. To overcome these discrepancies, I suggest that dephasing plays an important role in determining the transport properties of SAM-MJs. I will present a calculation approach that inherently takes into account in-plane dephasing in the electron motion as it traverses the SAM plane. The approach describes well the two hallmarks of transport through SAM-MJs, namely the exponential decay of current with molecular length and the reduction of the current per molecule as compared to single-molecule junctions. Specifically, I will show that dephasing leads to an exponential decay of the current as a function of molecular length, even for resonant tunneling, where the fully coherent calculation shows little or no length-dependence of the current. The dephasing is also shown to lead to a substantial reduction of the current in a SAM-MJ as compared to the single molecule junction, in a realistic parameter regime, where the coherent calculation shows only a very small reduction of the current. Finally, I discuss the effect of dephasing on more subtle transport phenomena such as the conductance even-odd effect and negative differential resistance.

**B04-11**

First principles electron transport through Au-benzenedithiol-Au molecular junctions with self-energy corrections

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Au-1,4-benzenedithiol(BDT)-Au junctions are the classical single-
molecule electronic systems. Many experimental and theoretical studies have been performed to study transport properties through this system. Despite these efforts, a clear understanding of the measured spread in electronic conductance is lacking, and the agreement between experimental and theoretical results is still unsatisfactory. A well-known problem of conventional theoretical approaches based on density functional theory (DFT), is the underestimation of the tunnelling barrier for transport. However, more sophisticated many-electron methods are time-consuming to perform and converge. Here, we employ a self-energy corrected first principles DFT-based quantum transport method, DFT+Sigma, [1] to thoroughly investigate the electronic conductance and thermopower of Au-BDT-Au molecular junctions. The method is implemented in a scattering state transport code, SCARLET. [2] We consider different contact geometries and compare our calculated results with existing experimental values. By overcoming the limitations of conventional DFT-based methods and high cost of many-electron methods, we obtain a better understanding of electronic transport characteristics of these junctions and proceed towards potential realistic realizations of these molecular-scale devices.


**B04-12 Invited**

Two Dimensional Electron Gas in Free Space: a First Principles Design

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Nearly free electron (NFE) states are widely existed on atomically smooth surfaces in two-dimensional materials. Since they are mainly distributed in free space, these states can in principle provide ideal electron transport channels without nuclear scattering. Unfortunately, NFE states are typically unoccupied, and electron doping is required to shift them toward the Fermi level and, thus, to be involved in electron transport. Instead of occupying these NFE states, it is more desirable to have intrinsic nucleus-free two-dimensional electron gas in free space (2DEG-FS) at the Fermi level without relying on doping. Inspired by a recently identified electrode material, we suggest that Ca$_2$N monolayer should possess such a 2DEG-FS state, which is then confirmed by our first-principles calculations. Phonon dispersion in Ca$_2$N monolayer shows no imaginary frequency indicating that the monolayer structure is stable. A mechanical analysis demonstrates that Ca$_2$N bulk exfoliation is feasible to produce a freestanding monolayer. However, in real applications, the strong chemical activity of 2DEG-FS may become a practical issue. It is found that some ambient Ca$_2$N bulk exfoliation is feasible to produce a freestanding monolayer. A mechanical analysis demonstrates that Ca$_2$N monolayer should possess such a 2DEG-FS state, which is then confirmed by our first-principles calculations. Phonon dispersion in Ca$_2$N monolayer shows no imaginary frequency indicating that the monolayer structure is stable. A mechanical analysis demonstrates that Ca$_2$N bulk exfoliation is feasible to produce a freestanding monolayer. Nevertheless, in real applications, the strong chemical activity of 2DEG-FS may become a practical issue. We extend the simple and efficient lowest order expansion (LOE) [1] for inelastic electron tunneling spectroscopy (IETS) to include variations in the electronic structure on the scale of the vibration energies. This enables first-principles calculations of IETS line shapes for molecular junctions close to resonances [2] and band edges. We demonstrate how this is relevant for the interpretation of experimental IETS using both simple models and first-principles simulations [2].


**B04-14 Invited**

Graphene Nanoribbon Junctions: Perfect Molecular Wire + Spin Valve

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For molecular wires to be useful in atomic scale circuitry, it is desirable that they exhibit a large conductance that does not decay with wire length. We predict, using first principles density functional theory transport calculations, that armchair graphene nanoribbon (AGNR) wires, connected by transverse zigzag edges to wider AGNR electrodes, can exhibit anomalous resonant transmission peaks close to $E_{\text{Fermi}}$, that are nearly independent of wire length. These peaks arise because the zigzag edge state at the interface can naturally and perfectly delocalize into the middle AGNR. The invariance of the peaks with wire length stems from the zigzag edges absorbing most of the wavefunction renormalization required when the wire length changes.[1] Furthermore, because the zigzag edge states are spin-polarized, these perfect transmission channels can be turned on or off with a magnetic field, suggesting promising applications as spin valves and transistors. In light of recent experimental advancements in bottom-up synthesis of atomically precise, several-nanometer-long AGNRs and AGNR junctions, our results pave the way for promising applications of AGNRs in nano-electronics and nano-spintronics.