O 78: Frontiers of Electronic Structure Theory: Many-Body Effects on the Nano-Scale V

Time: Thursday 10:30–13:15   Location: MA 004

Invited Talk   O 78.1   Thu 10:30   MA 004
Interaction and Correlation Effects in Quasi Two-dimensional Materials — •Steven G. Louie — Physics Department, University of California at Berkeley, and Lawrence Berkeley National Lab, Berkeley, CA 94720 USA

Experimental and theoretical studies of atomically thin quasi two-dimensional materials and their nanostructures have revealed that these systems can exhibit highly unusual behaviors. Owing to their reduced dimensionality, these systems present opportunities for manifestation of concepts/phenomena that may not be so prominent or have not been seen in bulk materials. Symmetry and many-body interaction effects often play a critical role in shaping qualitatively and quantitatively their properties. In this talk, we present some theoretical studies on graphene as well as other quasi-2D systems such as monolayer and few-layer transition metal dichalcogenides (e.g., MoS2, MoSe2, WS2, and WSe2) and metal monochalcogenides (such as GaSe and FeSe). Several quantum phenomena are discussed, including novel and dominant exciton effects, tunable magnetism, electron supercollimation by disorder, unusual plasmon behaviors, and possible enhanced superconductivity in some of these systems. We investigate their physical origins and compare theoretical predictions with experimental data.

O 78.2   Thu 11:00   MA 004
Screening of the Coulomb interaction in two-dimensional semiconductors: The case of transition metal dichalcogenides — •Ersoy Sasioglu, Christoph Friedrich, and Stefan Blügel — Peter Grünberg Institut and Institute forAdvanced Simulation, Forschungszentrum Jülich and JARA, 52425Jülich, Germany

Experimentally determined large exciton binding energies and nonhydrogenic Rydberg series in monolayer transition metal (TM) dichalcogenides indicate a long-range behavior of the Coulomb interaction. By means of first-principles calculations in conjunction with the random-phase approximation [1,2] within the FLAPW method [3] we study screening of the Coulomb interaction in two-dimensional semiconducting TM dichalcogenides MX_2 (M=Cr, Mo, W; X=S, Se). We show that the screening in these systems deviates substantially from the bulk behavior, i.e., the short-range interaction is strongly screened, while the long-range interaction is anti-screened. This unconventional screening reduces the gradient of the Coulomb interaction giving rise to weak correlation effects, which explains the experimentally observed large exciton binding energies as well as the success of the one-particle density functional theory in the description of the electronic structure of these systems. This work has been supported in part by DFG-FOR-1346.
[1] C. Friedrich *et al.*, Phys. Rev. B. **81**, 125102 (2010).
[2] E. Şaşıoğlu *et al.*, Phys. Rev. B **83**, 121101(R) (2011).
[3] www.flapw.de

O 78.3   Thu 11:15   MA 004
Ultra-fast transient absorption spectra of monolayer MoS2 by first principle — •Margherita Marsili[1], Deborah Prezzi[1], Davide Sangalli[2], and Andrea Marini[2] — [1]CNR Istituto di Nanoscienze S3, Modena, Italy — [2]CNR ISM, Montelibretti, Italy

We compute ultrafast transient absorption spectra of MoS2 monolayers by employing a novel approach which combines density-functional and non-equilibrium Green's function theories. This approach allows the description of pump-probe optical experiments where the system is excited by an ultrashort laser pulse, and the variation of the optical response is probed at different time delays, thus providing a wealth of information on the fundamental physics of the relaxation processes. The case of monolayer MoS2 is extremely challenging due to the interplay of excitonic, electron-phonon and spin-orbit coupling effects. We describe the excitation of the MoS2 electronic system and follow the subsequent dynamics using a fully non-collinear spin formulation of the theory, including excitonic effects. The results are compared with experimental pump-probe data.

O 78.4   Thu 11:30   MA 004
Plasmon and exciton dispersion in two dimensions — •Pier Luigi Cudazzo — LSI Ecole Polytechnique and ETSF, Palaiseau, France

Understanding the electronic properties of 2D materials requires the investigation of their elementary excitations that dictate their optical and transport properties. Using state-of-the-art Green's function many body approach we present a first principle study of the collective excitations (namely excitons and plasmons) in 2D materials. In particular from the evaluation of the dielectric function we investigated the exciton dispersion in graphane and hBN and the plasmon dispersion in metallic TMDs[1-3]. From our results we provide an exact analytic form of the two-dimensional screened potential. In contrast to 3D systems where the macroscopic screening can be described by a static dielectric constant in 2D systems the macroscopic screening is non local (q-dependent) showing a logarithmic divergence for small distances and reaching the unscreened Coulomb potential for large distances[4].
[1] P. Cudazzo, et. al. New J. Phys. 15 125005 (2013). [2] P. Cudazzo, et. al. (in preparation) [3] P. Cudazzo, et. al. Phys. Rev. Lett. 104 226804 (2010). [4] P. Cudazzo, et. al. Phys. Rev. B 84 085406 (2011).

O 78.5   Thu 11:45   MA 004
Origin of metallic edge states in transition-metal-dichalcogenide nanostructures — •Marco Gibertini and Nicola Marzari — Theory and Simulation of Materials (THEOS) and National Center for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, Switzerland

The existence of metallic edge states in transition-metal-dichalcogenide nanostructures has been reported both in the experimental and theoretical literature. Such nanostructures include for instance triangular islands and zigzag nanoribbons. Nonetheless, a thorough understanding of the mechanism giving rise to metallic states at the edge of such bulk insulating materials is still missing. Here we suggest a possible origin of such states and support our findings with first-principles density-functional-theory simulations. The key observation is that transition metal dichalcogenides like MoS_2 display a finite *formal* polarization that induces a charge reconstruction with the appearance of free carriers at the edges. We also suggest possible innovative applications in nanoelectronics and solar-energy devices.

O 78.6   Thu 12:00   MA 004
Starting-point dependence in the Bethe-Salpeter equation: example of rutile TiO_2 — •Olga Turkina, Ute Werner, Dmitrii Nabok, and Claudia Draxl — Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, Zum Großen Windkanal 6, D-12489 Berlin, Germany

Many-body perturbation theory, combining the GW approach and the Bethe-Salpeter equation (BSE), is a powerful tool for the description of one- and two-particle excitations. However, employing density functional theory as a starting point for these methods may lead to results that depend on the choice of the exchange-correlation (xc) functional. This starting-point dependence is well known for the G_0W_0 approximation. Such dependence is expected to occur also for BSE calculations, however, has not yet been investigated. With the example of rutile TiO_2, we show that this is, indeed, the case. We employ two different xc functionals: the generalized gradient approximation (PBE) and a hybrid functional (PBE0'), combining a fraction of 25% exact exchange with PBE. The electronic structure is calculated using the G_0W_0 approximation. The BSE is solved to obtain the optical absorption spectra. These are analyzed with regard to the influence of eigenvalues, wave functions, and screening as originating from different xc functionals.

O 78.7   Thu 12:15   MA 004
Efficient exchange-correlation kernels for the description of excitonic effects in solids — •Santiago Rigamonti[1,4], Silvana Botti[2,4], Valérie Veniard[3,4], Claudia Draxl[1,4], Lucia Reining[3,4], and Francesco Sottile[3,4] — [1]Physics Department, Humboldt-Universität zu Berlin, Germany — [2]Friedrich-Schiller-Universität Jena, Institut für Festkörpertheorie und -optik — [3]Laboratoire des Solides Irradiés, École Polytechnique, CNRS, CEA-DSM, F-91128 Palaiseau, France — [4]European Theoretical Spectroscopy Facility (ETSF)

One of the major challenges for time-dependent density-functional the-

ory is the accurate and efficient description of excitonic effects in solids, captured by the exchange-correlation (xc) kernel. In a recent empirical approach, the so-called "bootstrap" kernel has been proposed. Due to its high efficiency and some promising results [1] it appeared indeed interesting. In this work, we find a physically motivated derivation for it, opening the way to understand its weaknesses and to propose a new xc kernel. Our kernel is both simpler and more reliable, as confirmed by our numerical results [2]. We also propose a simple method to estimate exciton binding energies from the dielectric functions computed in the random-phase approximation alone. This method makes the approach accessible to a wide range of scientists.

[1] S. Sharma, J. K. Dewhurst, A. Sanna, and E. K. U. Gross, *Phys. Rev. Lett.* **107**, 186401 (2011).
[2] S. Rigamonti, S. Botti, V. Veniard, C. Draxl, L. Reining, and F. Sottile, *submitted*.

O 78.8   Thu 12:30   MA 004

Excitonic effects in many-body calculations — ●Matteo Gatti[1,2], Igor Reshetnyak[1], Giorgia Fugallo[1], Pierluigi Cudazzo[1], Francesco Sottile[1], and Lucia Reining[1] — [1]LSI, CNRS-Ecole Polytechnique and ETSF, Palaiseau, France — [2]Synchrotron Soleil, Gif-sur-Yvette, France

The Bethe-Salpeter equation (BSE) is the state-of-art approach to calculate the absorption spectra of a large variety of materials [1]. Here we show that that the BSE is a powerful and accurate method also for the calculation of the exciton dispersion [2-4] (i.e. the exciton energy as a function of the momentum q carried by the electron-hole pair), and of the off-diagonal elements of the dielectric function in reciprocal space $\epsilon_{G,G'}(q,\omega)$ [5]. On the one hand, this allows the ab initio simulation of spectra measured by Electron Energy-Loss Spectroscopy (EELS) and Inelastic X-ray Scattering (IXS), including its Coherent version (CIXS), well beyond the optical limit $q\to0$. On the other hand, this opens the door to the calculation of spectral functions [6-8] using the cumulant expansion for the Green's function G with a screened Coulomb interaction W that includes excitonic effects beyond the random-phase approximation employed in the GW approximation.

[1] G. Onida, *et al.*, Rev. Mod. Phys. **74**, 601 (2002). [2] M. Gatti and F. Sottile, Phys. Rev. B **88**, 155113 (2013). [3] P. Cudazzo, *et al.*, Phys. Rev. B **88**, 195152 (2013). [4] G. Fugallo, *et al.*, unpublished. [5] I. Reshetnyak, *et al.*, unpublished. [6] M. Guzzo, *et al.*, Phys. Rev. Lett. **107**, 166401 (2011). [7] M. Gatti and M. Guzzo, Phys. Rev. B **87**, 155147 (2013). [8] M. Guzzo, *et al.*, Phys. Rev. B **89**, 085425

(2014).

O 78.9   Thu 12:45   MA 004

Efficient parameter-free calculation of absorption spectra for insulators, semiconductors and metals from time-dependent current DFT — ●Arjan Berger — LCPQ - IRSAMC, Université de Toulouse III - Paul Sabatier, CNRS, Toulouse, France and European Theoretical Spectroscopy Facility

In this work we show that with a simple dynamical kernel we can obtain good absorption spectra from time-dependent current-density functional theory (TDCDFT) for insulators, semiconductors and metals. Our approach is fully parameter free since no artificial broadening parameter is used to match calculated and measured spectra. The cost of a calculation is equal to an RPA calculation. Moreover, our TD-CDFT approach scales better with system size than standard TDDFT implementations.

O 78.10   Thu 13:00   MA 004

Optical excitations in MoS$_2$ within ab-initio many-body perturbation theory — ●Matthias Drüppel, Peter Krüger, and Michael Rohlfing — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität, 48149 Münster, Germany

The transition metal dichalcogenides (TMDC), and MoS$_2$ as its most prominent member, open the door to a field of two dimensional atomically thin semiconductors which offer rich physics.

The state of the art theoretical description of electronic excitations in these materials starts with density-functional calculations (DFT), followed by the GW method in combination with a solution of the Bethe-Salpeter equation. The *converged* calculation (especially with respect to the size of the basis, i.e. number of plane waves, and k-meshs) of the last two steps has shown to be numerically extremely challenging.

We apply the efficient LDA+GdW [1] approach to the excited states which enables us to describe electronic excitations in MoS$_2$ at substantially lower numerical cost. In the LDA+GdW approximation the quasiparticle self-energy corrections to the LDA-DFT energies result from the difference between the correct screening (semiconducting) and hypothetical metallic screening.

This approach sets us in a position where more atoms per unit cells can be treated, e.g., for defects and for MoS$_2$ on substrates.

[1] M. Rohlfing, Phys. Rev. B. **82**, 205127 (2010)