Absorption and hot carrier recombination in van der Waals Materials

Pedro Venezuela,[1,2] Enésio Martinho Júnior,[1] César Perez Villegas,[1,3] Alexandre Reily Rocha

[1]Instituto de Física Teórica, São Paulo State University, São Paulo, Brazil

[2] Departamento de Física, Universidade Federal Fluminense, Niterói, Brazil

[3] Departamento de Ciencias, Universidad Privada del Norte, Lima 15434, Peru

Van de Waals materials comprise a class of layered materials that have attracted a great deal of attention. In the field of highly attractive for optoelectronics this is due to their scalability and thickness-dependent electrical and optical properties [1]. Within this class, transition metal dichalcogenides (TMDC) are highly attractive for various applications including photovoltaics. While there is considerable work dealing with single layer TMDCs, for the few-layer case is somewhat neglected.

As a first step, here we will present the electronic and optical properties of bulk 2H group VIB TMDs. We performed many-body perturbation theory (MBPT) simulations to compute the quasiparticle bandstructure within G_0W_0, and subsequently solve Bethe Salpeter equation for the optical absorption including electron-hole correlations [2,3,4] using the Yambo code. Our results correlate extremely well with existing experimental results, particularly for the excitonic peaks. We show that, due to the symmetry of the bands, the first exciton present in all these systems is dark, occurring in the vicinity of K point, whereas the brighter excitons occur in H-K high-symmetry direction, with transitions between the VBM and CBM+1, and strong interlayer component. Furthermore, using that information, we estimate the theoretical photovoltaic performance of these group VIB TMDs by calculating the short-circuit current, open-circuit voltage and power conversion efficiency, presenting values higher than semiconductors of similar width [6].

In the second part of this talk we will discuss other important features requied for a good for photovoltaic applications. In particular, using of *ab initio* many-body perturbation theory, including the spin–orbit interaction, we investigate the photocarrier generation and dynamics in α-Tellurene [6]. We show that photoexcited electrons are mainly generated in the near-infrared range, forming excitons that are strongly bound, compared to its bulk counterpart. We also explore the role of the electron–phonon and electron-electron interactions in the relaxation of charged carriers. We find that the electronic states in the first conduction band minimum couple weakly with phonons, yielding longer hot electron lifetimes, and mean free paths up to 37 nm. We also show that the extraction of hot holes may result in a challenging task as these carriers possess sub-3 nm mean free paths. We

finally estimate that 1-nm-thick α-Te provides a short-circuit current density of 6.7 mA/cm^2 and a maximum power conversion efficiency of 4.4%, comparable to other layered materials, which highlights its potential for efficient photovoltaic device development.

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