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Unveiling the Roles of Excitons with Ab Initio Principles Calculations for Mono- and Multi-Layered Black Phosphorus.

Since its recent synthesis, Black Phosphorus (BP) has been attracting interest due to its peculiar properties: unlike the well-known graphene, BP (as shown in Figure 1) is a semiconductor, possessing a band gap that can be be tuned by the number of stacking layers (going from 0.3 eV for the bulk BP, to approximately 2.0 eV for the monolayer configuration)[1-2]. In this work we study the electronic and optical properties of this material, and its various stacking configurations, using Density Functional Theory and the GW-BSE approximation method. This is required since DFT alone, in the most common exchange-correlation approximations used, does not provide good description of the electronic band structure. The GW-BSE approach is also necessary to comprehend the optical properties of the material and study the role of excitons [2]. Precise exciton binding energy for a system is a key measure of excitonic effects. These effects can involve third harmonic generation (THG)[3], where knowing the electronic spectra for different number of layers can explain the resonantly increased optical frequency conversion. The computational work is carried out using the QuantumESPRESSO software package alongiside BerkeleyGW code, which can compute the excitonic spectrum. Results for energy gap, optical gap and excitonic spectrum agree with those obtained by other work [1-3,5], and are able to explain recent finding for THG in the material [3]. Our preliminary results, as shown in Figure 2, show that for bulk BP the absorption spectrum is well described without excitonic effects. On the other side, we demonstrated that a more accurate and precise description of the absorption spectrum of monolayer BP is obtained when excitonic effects are accounted for in the GW-BSE approach.

References

[1]: F.Ferreira, R.M. Ribeiro - GW approxmation and BSE calculations on phosphorene. (2017)
[2]: Jin-Ho Choi, Ping Cui, Haiping Lan, Zhenyu Zhang - Linear scaling of the exciton binding energy versus the bandgap of two-dimensional materials. PRL 115, 066403 (2015)
[3]: Manuel J. L. F. Rodrigues, Christiano J. S. de Matos, Yi Wei Ho, Hélder Peixoto, Rafael E. P. de Oliveira, Hsin-Yu Wu, Antonio H. Castro Neto, and José Viana-Gomes - Resonantly Increased Optical Frequency Conversion in Atomically Thin Black Phosphorus. adma 201603119 (2016)
[4]: L.Li, Y.Yu, G.J. Ye, Q. Ge, X. Ou, H. Wu, D. Feng, X. H. Chen, Y. Zhang. Nat Nano 9, 372 (2014)
[5]: Vy Tran, Ruixiang Fei, Li Yang - Quasiparticle energies, excitons and optical spectra of few-layer black phosphorus. 2D Mater.2 044014 (2015)

Figures

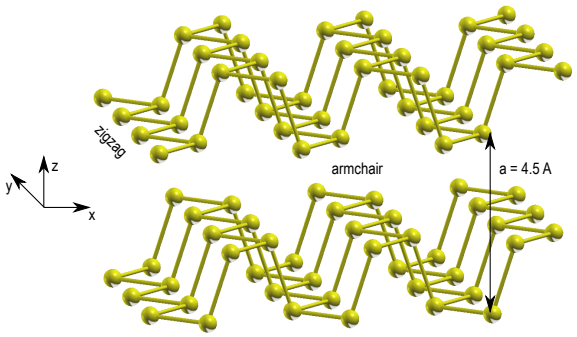

Figure 1: Structure of Black Phosphorus in 3D.

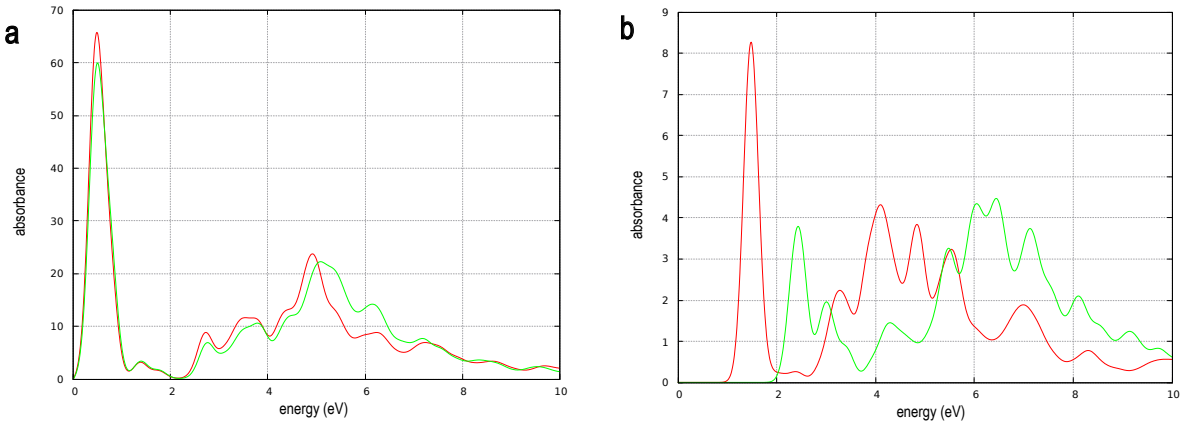

Figure 2: GW-BSE calcutated optical spectrum for (a) bulk and (b) monolayer Black Phosphorus. The absorption spectrum for incident light is along the armchair direction. Green curves represent the energy spectrum when excitons are not taken into account, red curves represent the absorption spectrum when excitons are accounted for in the GW-BSE approach.