Momentum-resolved dielectric response of free standing black phosphorus down to the monolayer

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Anisotropy and layer number drive the semiconducting properties of thin black phosphorus (BP), that offers promising perspectives in various fields such as electronics and photonics. However, the fast photooxidation in ambient condition, coupled to a high sensitivity to quantum confinement and dielectric environment in ultrathin BP, make very difficult the investigations on its intrinsic optical properties [1]. Further, as screening effects may strongly affect electronic and spectroscopic properties of 2D materials, it is highly desirable to investigate intrinsic properties of free-standing layers.

We have shown that Angular resolved Electron energy loss spectroscopy implemented in Transmission Electron Microscopy (ar-EELS-TEM) offers a unique way to investigate dielectric response of free-standing layers related to valence band and plasmon excitations with the advantage to get access to their q dispersion and their symmetry properties [2]. By combining this technique with suitable ab initio calculations, we have studied the dielectric response of free-standing BP layers as a function of the number of layers.

We found optical bandgap values of 1.9 eV, 1.4 eV and 1.1 eV for the mono- bi- and trilayer respectively. Moreover, by combining our results with a simple variational model, we correlate the exciton energy with the dielectric screening. We hence demonstrate that the variations of the electronic gap are sizeably larger than the variations of the binding energy. Finally, we probe and analyze the volume and surface plasmons dispersion as a function of momentum for the 1-3 BP layers and bulk and highlight a deviation and linearization of the parabolic dispersion with strong anisotropic fingerprints [3].

References

[1] A. Favron, E. Gaufres et al Nature Mat. 14 (2015) 826.
[2] F. Fossard et al, Phys. Rev. B 96, 115304 (2017)
[3] E. Gaufres et al, in preparation (2019)

Figures

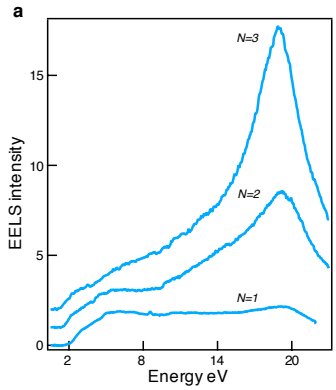

Figure 1: EELS spectra of suspended BP mono, bi and trilayers (N=1, 2, 3) showing the optical gap and the plasmon peak.

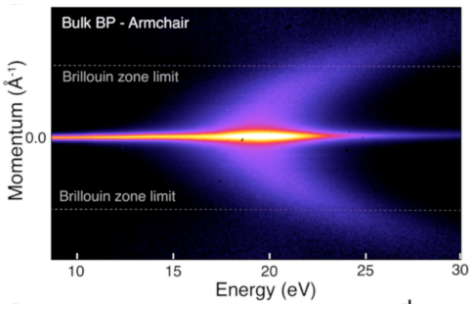

Figure 2: Mapping of the plasmon energy loss along the armchair direction of BP