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Supporting Information: Magneto-optical response of chromium trihalide monolayers: chemical trends

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Dielectric screening

In 2D materials, the bandgap and hence the dielectric screening determines the exciton binding energy at large extent. In Fig. 1a we have represented the exciton binding energies as a function of the bandgap and found a linear trend (also predicted in ref.[1]). We find an analogous trend in the dielectric screening. In Fig. 1b we show the head of the static dielectric screening, $\epsilon^{-1}_{0,0}(|\mathbf{q}|)$. We obtain that $\epsilon^{-1}_{\mathrm{Cl}} < \epsilon^{-1}_{\mathrm{Br}} < \epsilon^{-1}_{\mathrm{I}}$. Therefore the larger exciton binding energy is associated to a smaller dielectric screening.[2] For comparison we have included the screening of single-layer MoS_2, that has a binding energy of 0.5 eV.

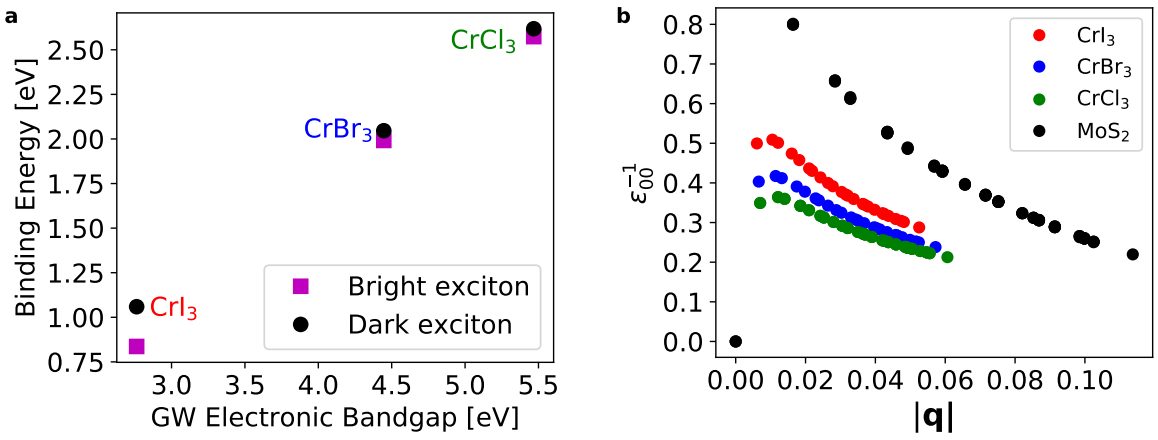

Figure 1: a. Excitonic binding energy of the bright and dark exciton as a function of GW bandgap. b. Head of the static dielectric screening matrix for the family of chromium trihalides and single-layer MoS_2.

Bandgap and Hubbard correction

The main effect of the Hubbard correction is the splitting of the $e_g - t_{2g}$ conduction bands. The bandgap increases only for U values smaller than 1 eV and it reduces for larger values, as shown in Fig. 2a. The main effect of the Hubbard correction is the splitting of the conduction band states e_g and t_{2g}.

Regarding the GW method and the Hubbard correction, Figure 2b shows the GW bandgap as a function of U, compared with the change in the DFT bandgap. Basically, the GW bandgap follows the same trend than the DFT one. The small difference can be attributed to the change of dielectric screening with U. In any case, the optical spectra does not change significantly with U within a reasonable range of values.

MOKE. Independent Particle Approximation

We show in Fig. 3 the Kerr spectra for the three chromium trihalides, with (BSE) and without (IP) excitonic effects. The general trend is a slight renormalization of the first and

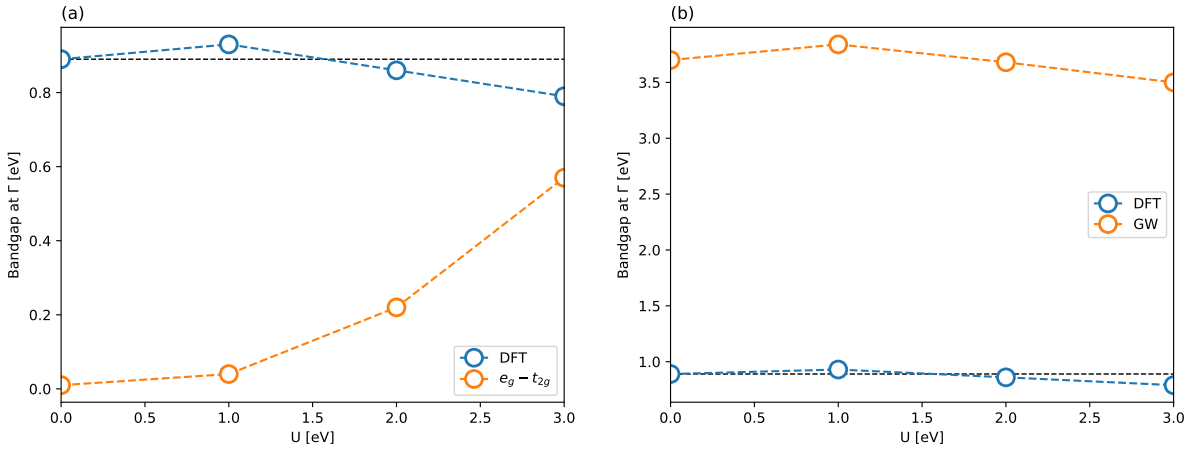

Figure 2: (a) Bandgap at Γ as a function of U. (b) Bandgap at Γ as a function of U using DFT and GW approximation.

second peak. The overall line shape of BSE and IP spectra are rather similar in the three cases.

References

(1) Jiang, Z.; Liu, Z.; Li, Y.; Duan, W. *Phys. Rev. Lett.* **2017**, *118*, 266401.

(2) Raja, A.; Chaves, A.; Yu, J.; Arefe, G.; Hill, H. M.; Rigosi, A. F.; Berkelbach, T. C.; Nagler, P.; Schüller, C.; Korn, T. et al. *Nature Communications* **2017**, *8*, 15251.

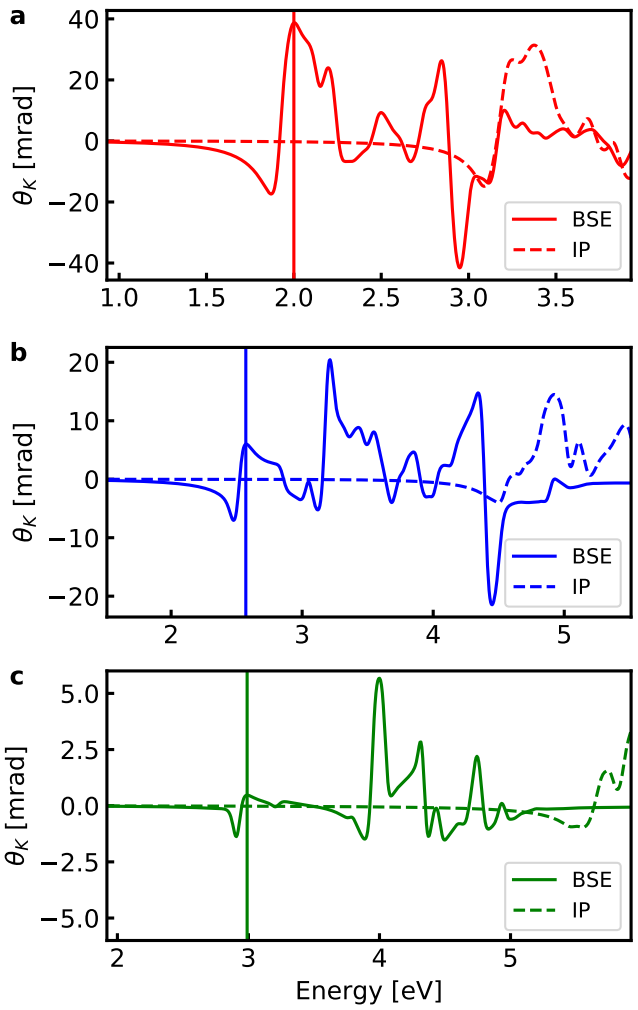

Figure 3: Kerr angle spectra with (solid lines) and without (dashed lines) excitonic effects of (a) CrI_3, (b) $CrBr_3$ and (c) $CrCl_3$.