Assessing G_0W_0 for d- and f-electron systems

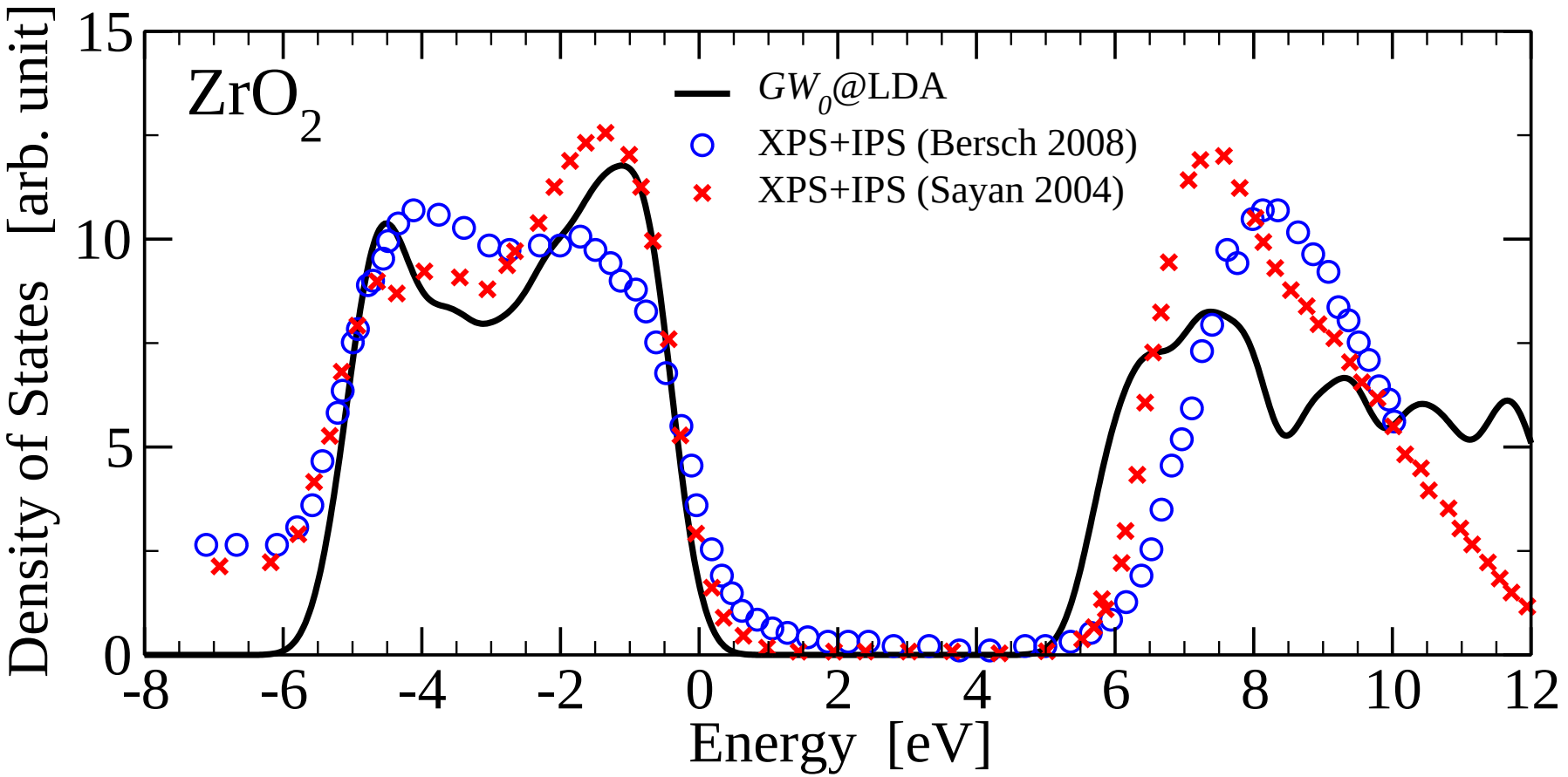
Selection of d- and f-electron systems

empty f	filled f	partially occupied d	partially occupied f
ZrO_2	HfO_2	MO (M=Ni, Mn, Fe, Co)	Ln_2O_3 (Ln=lanthanide series)

How well will G_0W_0 perform?

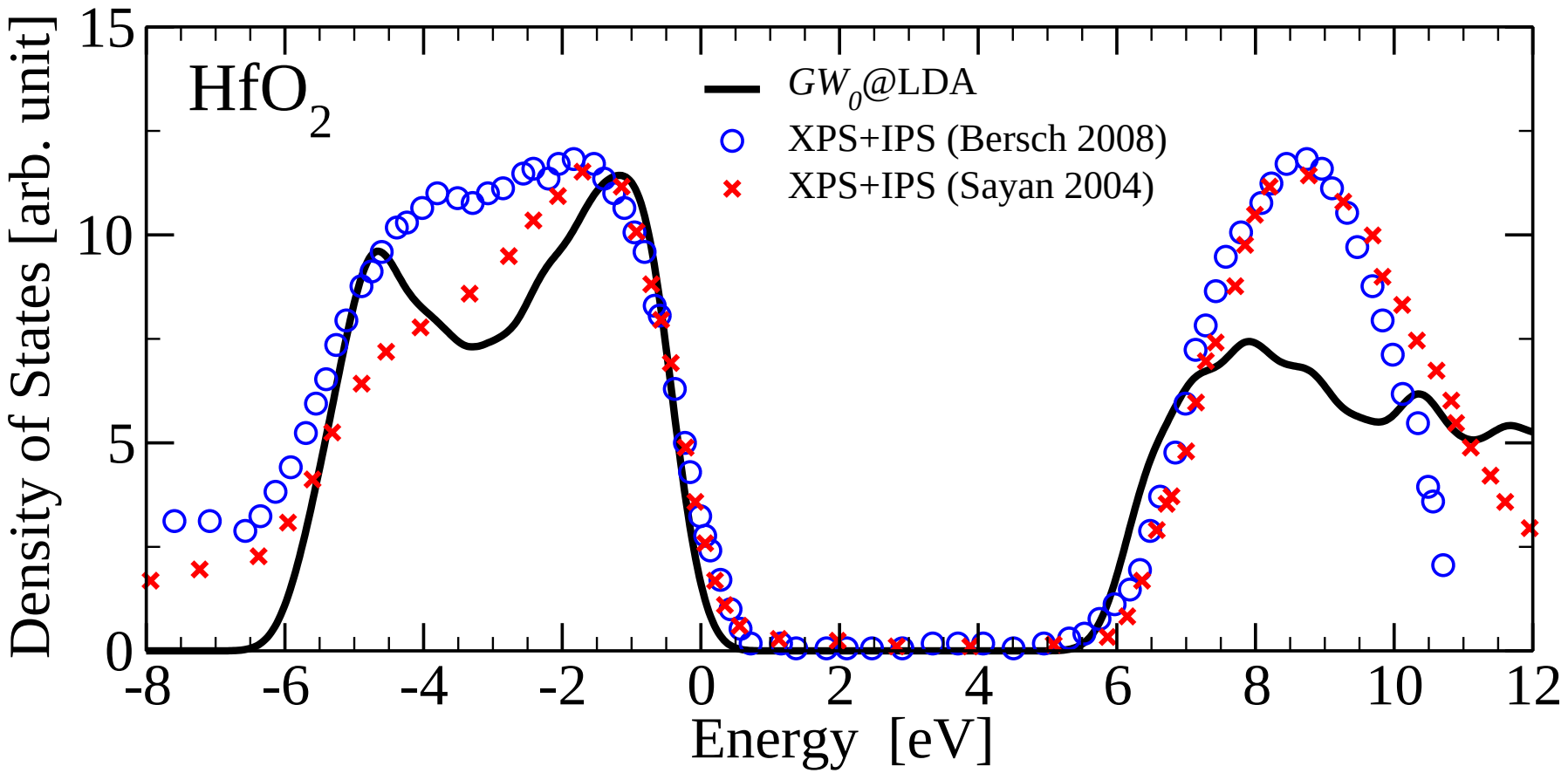
- exact exchange (but with DFT orbitals)
  - essential for localized electrons
- screening
  - essential for itinerant electrons
- answers from new LAPW-based *GW* code

Empty f-states – GW_0@LDA for ZrO_2



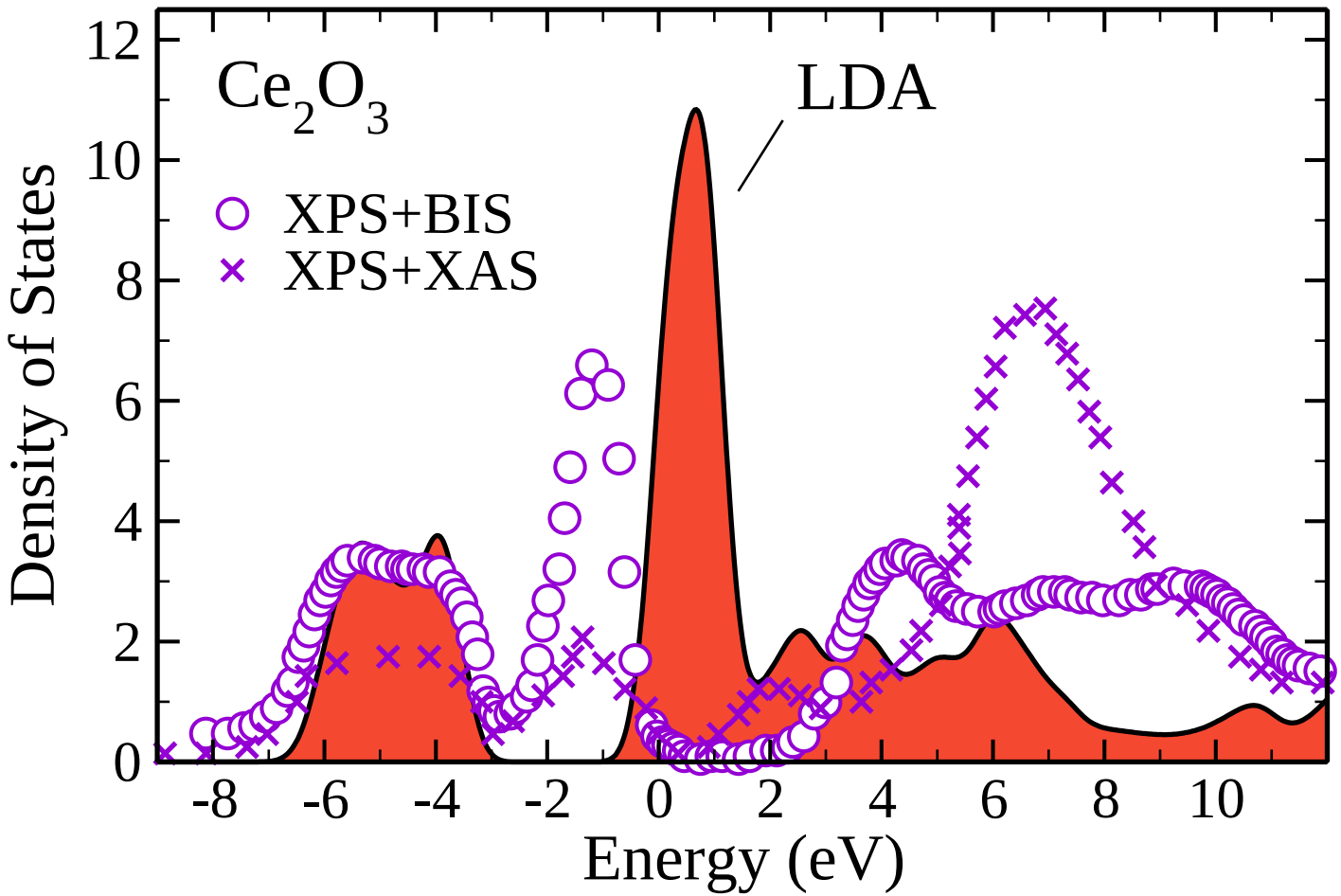
- GW_0@LDA agrees well with photoemission data

Filled f-states – GW_0@LDA for HfO_2



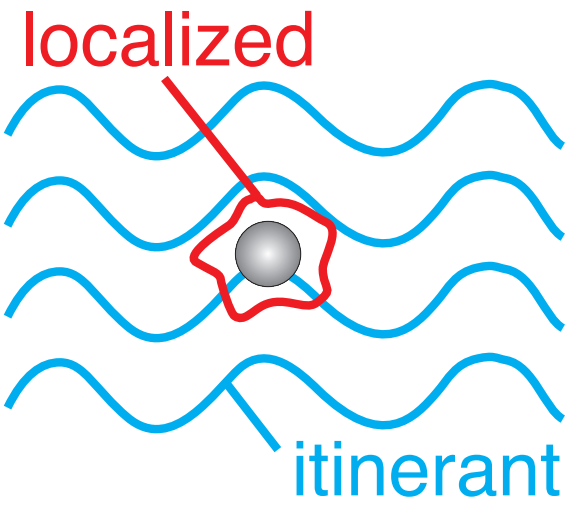
- GW_0@LDA agrees well with photoemission data

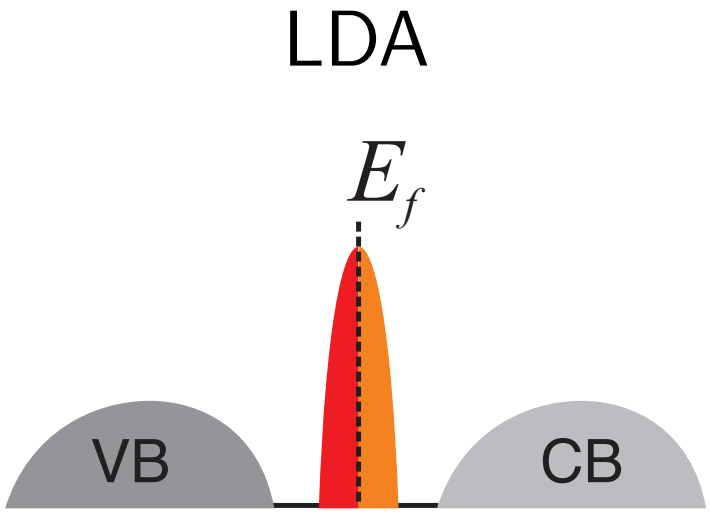
Ce_2O_3 - Failure of the LDA



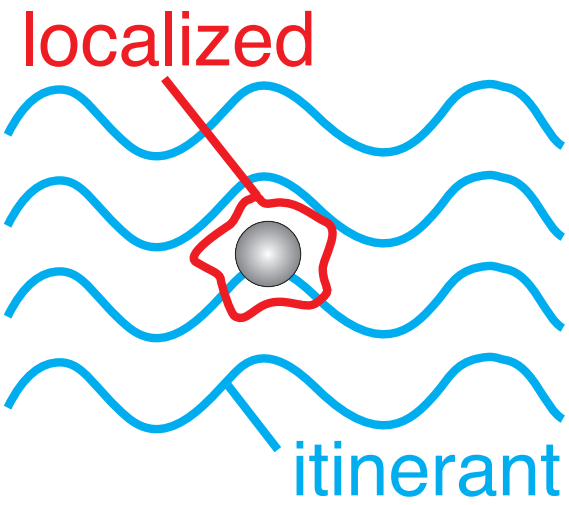
- LDA erroneously predicts metallic state

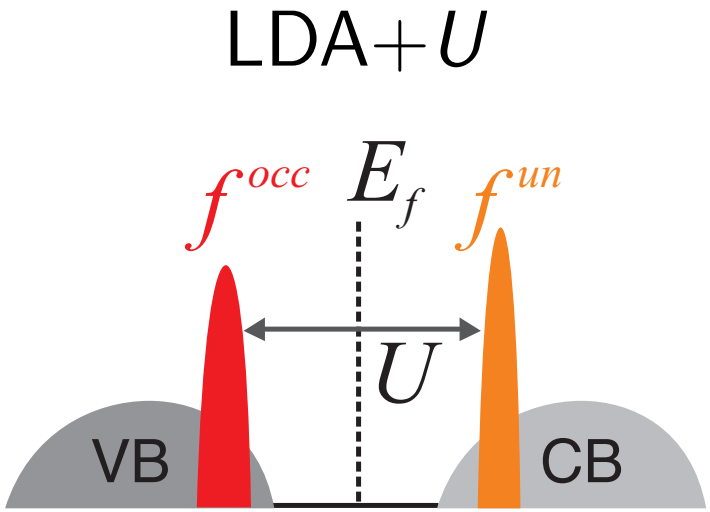
Ce_2O_3 - Localized vs Itinerant States





Ce_2O_3 - Localized vs Itinerant States



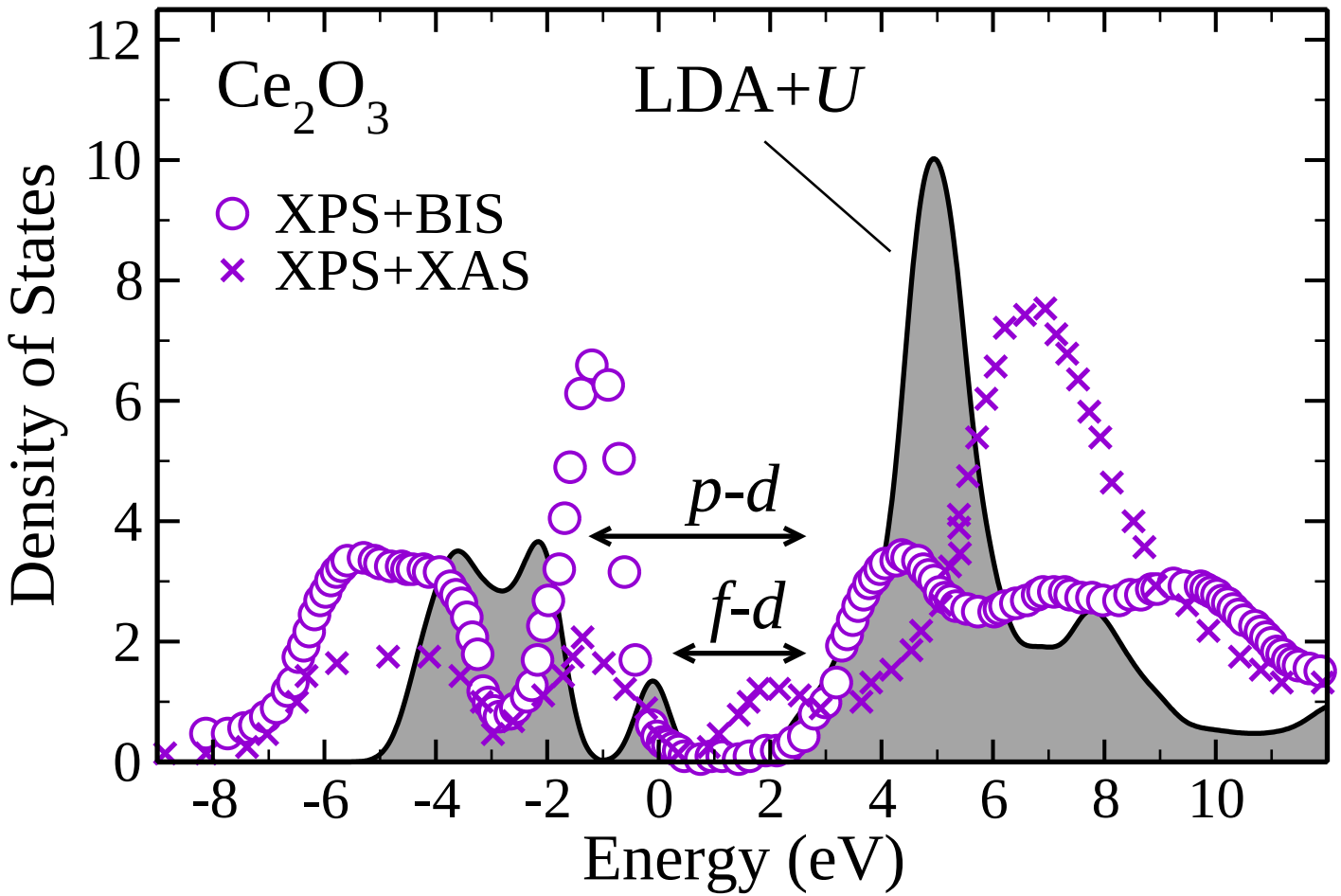


$$E^{\mathrm{LDA}+U}[\rho(\mathbf{r}), \hat{n}_I^a] = E^{\mathrm{LDA}}[\rho(\mathbf{r})] + E^{\mathrm{ee}}[\hat{n}_I^a] - E^{\mathrm{dc}}[\hat{n}_I^a]$$

E^{ee} : screened HF-like interaction between localized electrons

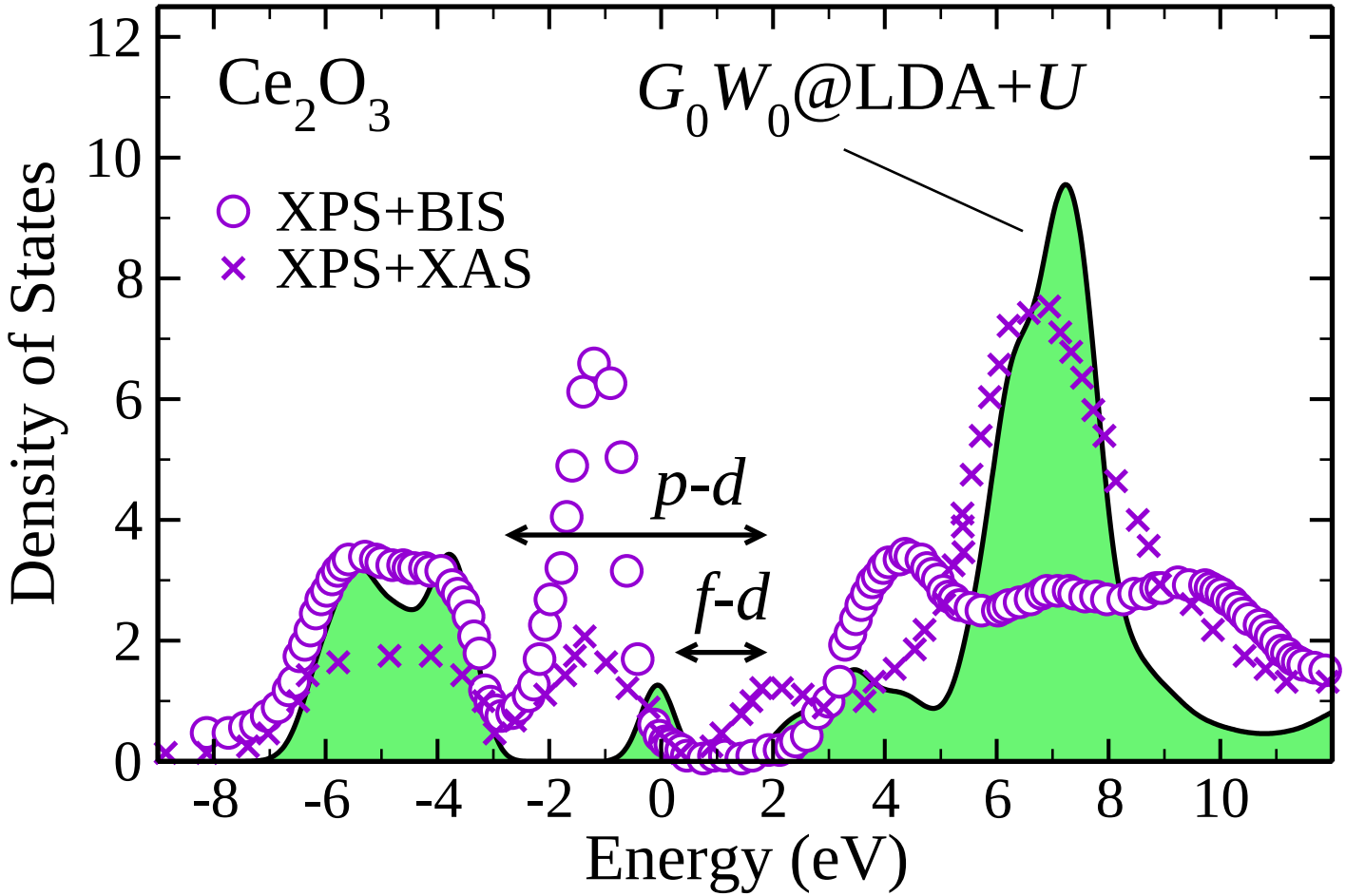
E^{dc} : double counting correction

Ce_2O_3 in LDA+U



- LDA+U splits f-peaks and opens gap

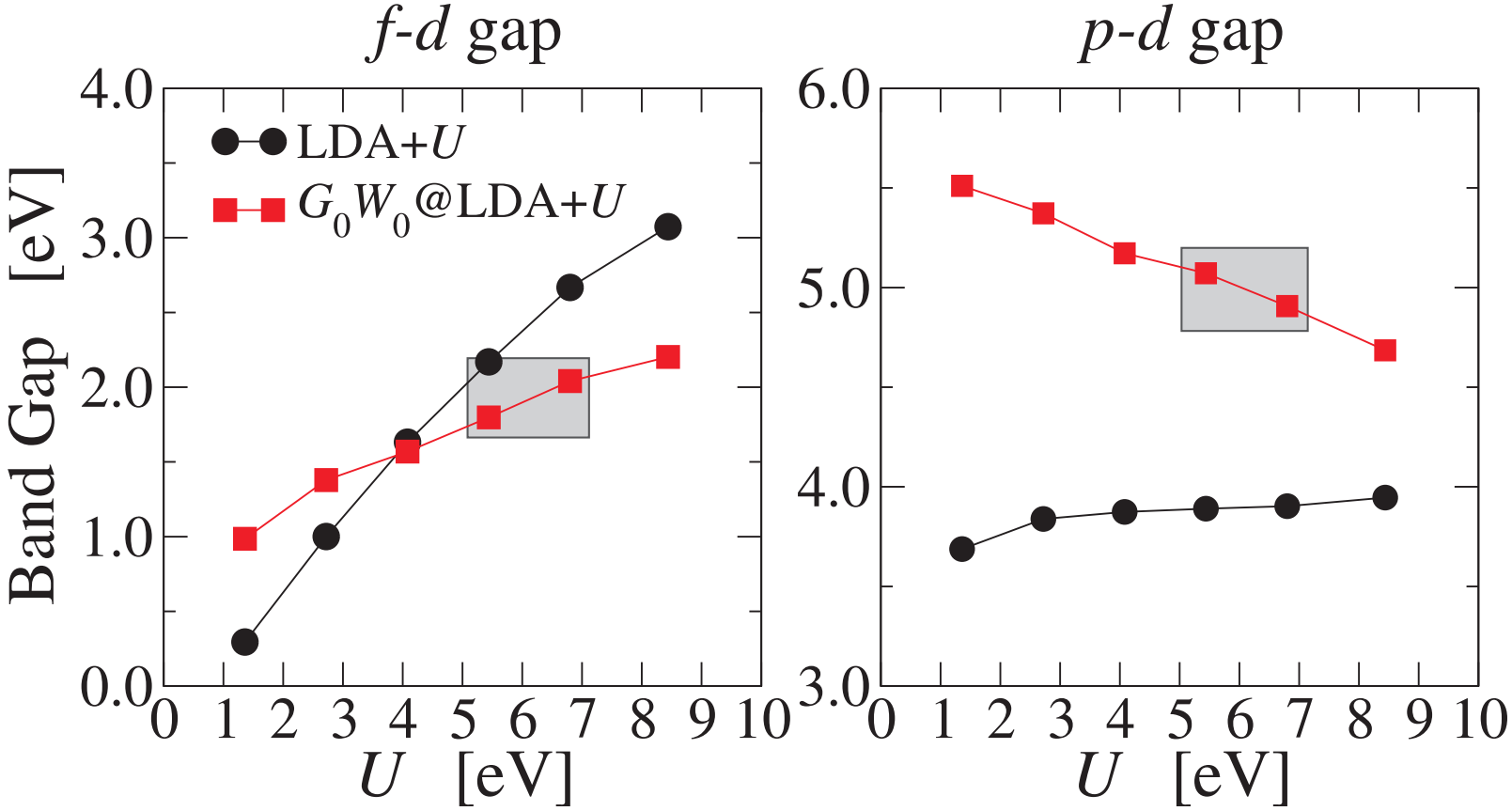
Ce_2O_3 in G_0W_0@LDA+U



- G_0W_0@LDA+U (U=5.4 eV) reproduces main peaks and gaps

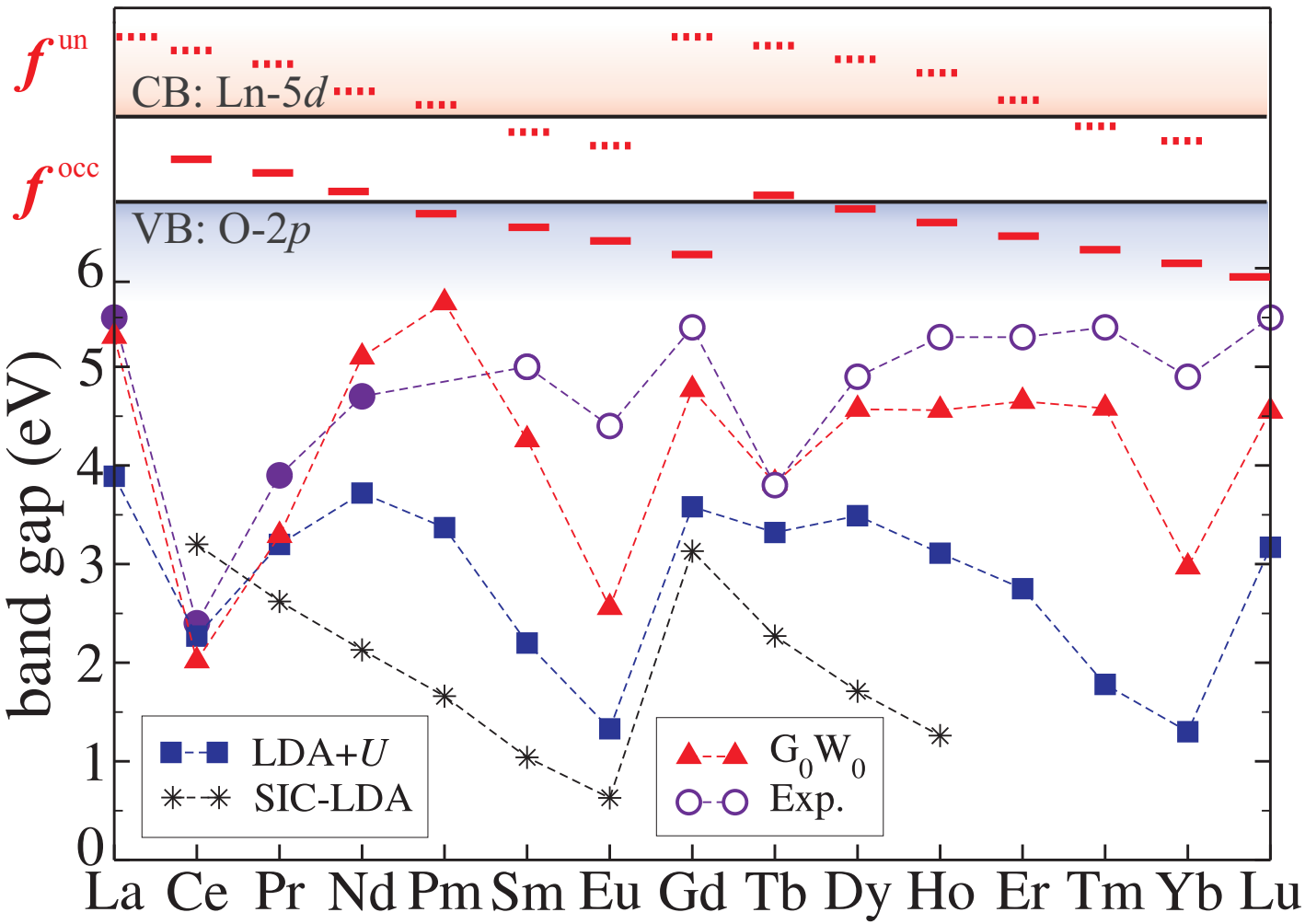
H. Jiang, *et al.*, *Phys. Rev. Lett.* **102**, 126403 (2009)

Ce_2O_3 – U-dependence in G_0W_0@LDA+U



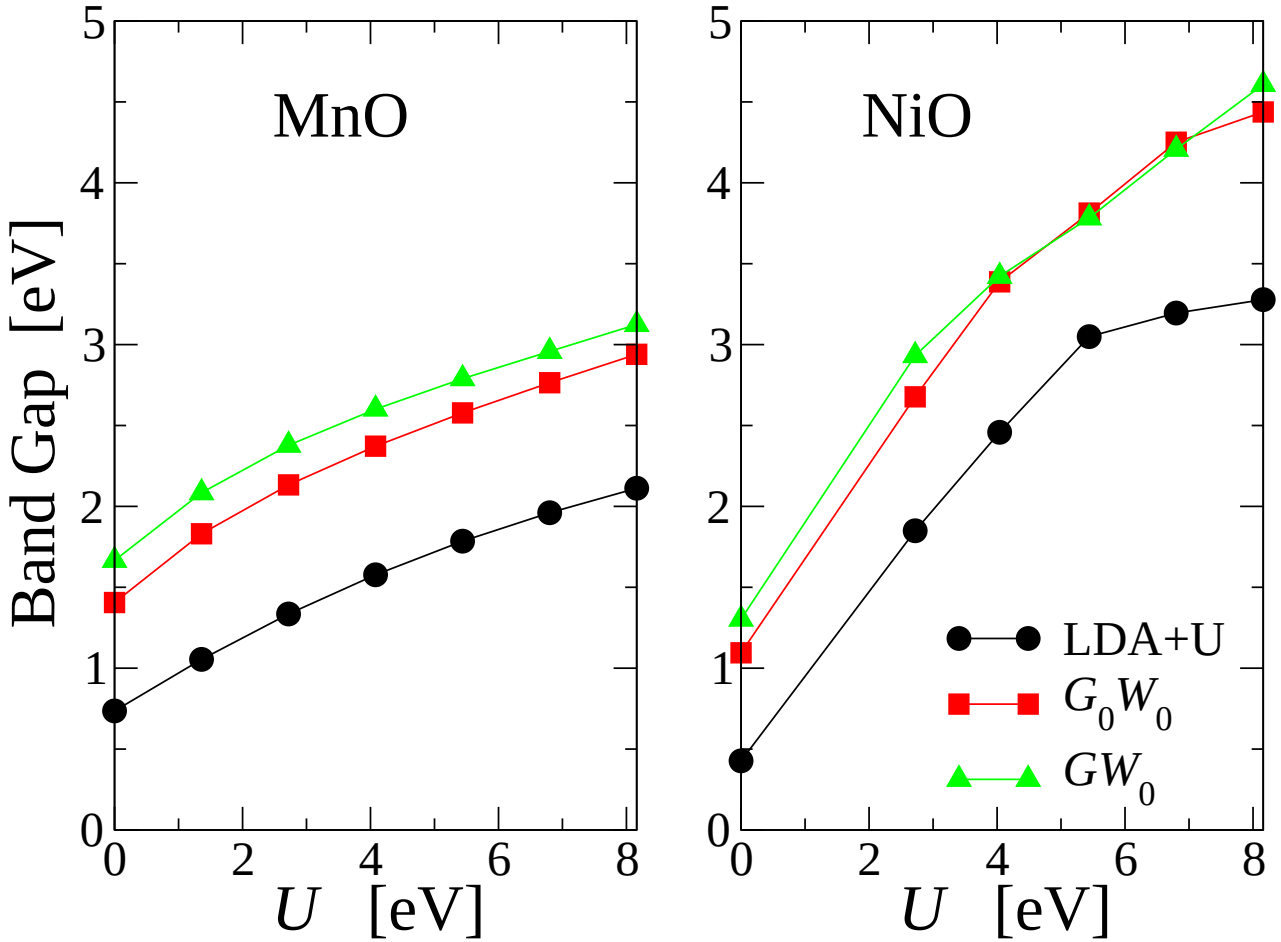
- G_0W_0@LDA+U: small U-dependence in "physical range"

Lanthanide Sesquioxide Series



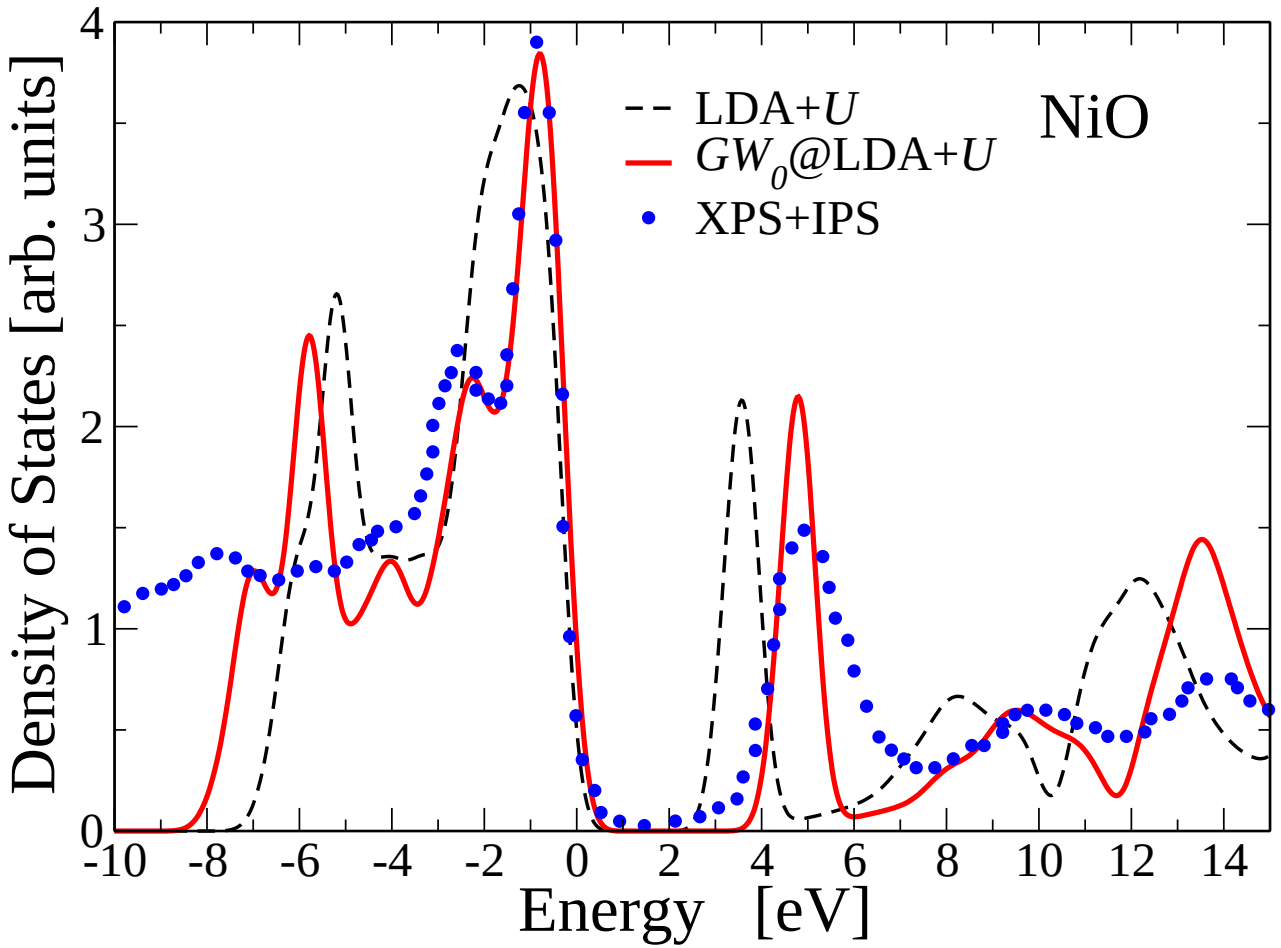
H. Jiang, *et al.*, *Phys. Rev. Lett.* **102**, 126403 (2009)

Transition metal oxides



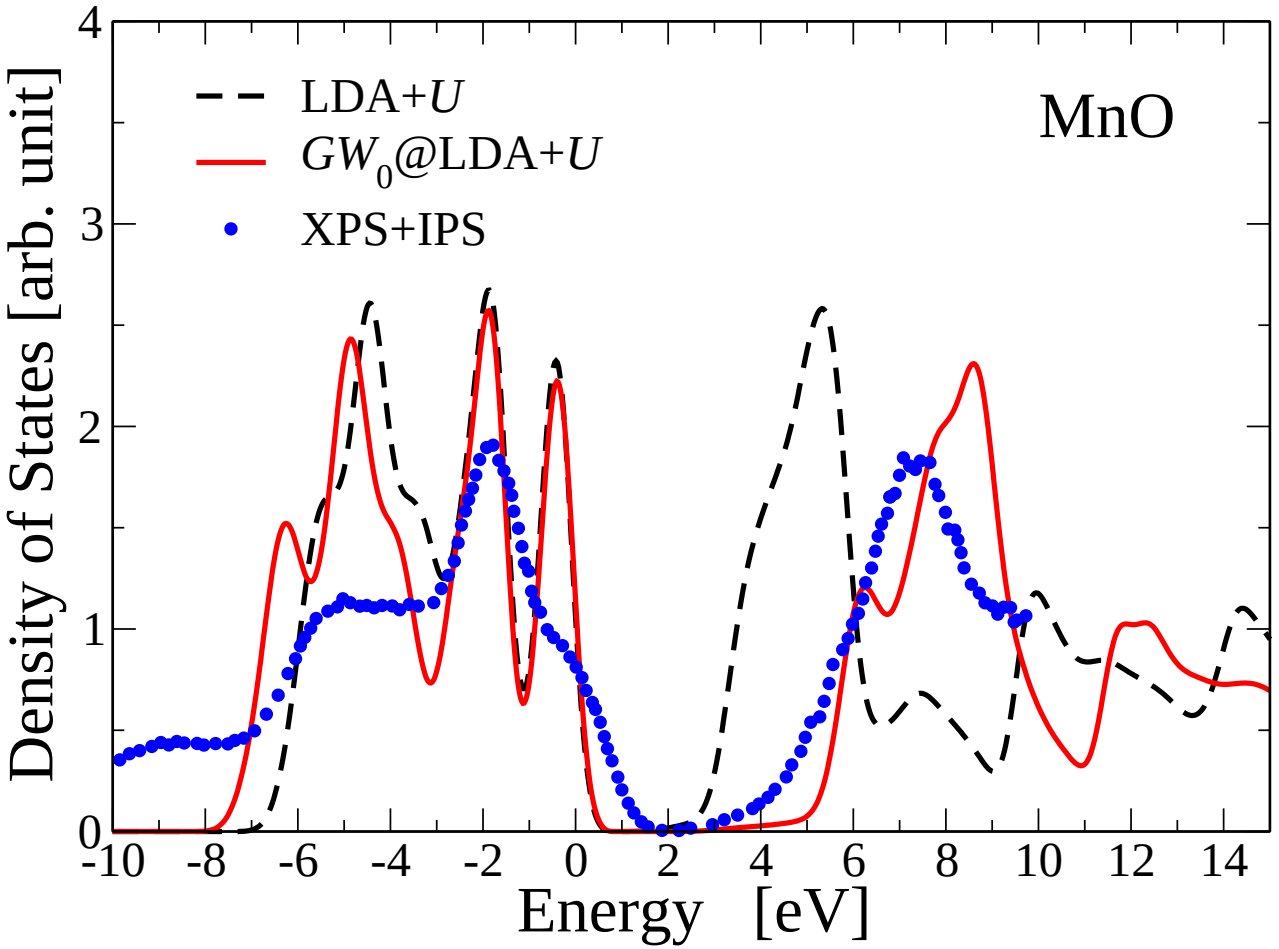
- strong U dependence in G_0W_0 and GW_0@LDA

Transition metal oxides – NiO



- XPS-IPS: G. A. Sawatzky *et al.* PRL **53**, 2339 (1984)
- U=5.2 eV, J=0.9 eV (from constrained DFT)

Transition metal oxides – MnO



- XPS-IPS: J. van Elp *et al.* PRB **44**, 1530 (1991)
- U=4.7 eV, J=0.8 eV (from constrained DFT)