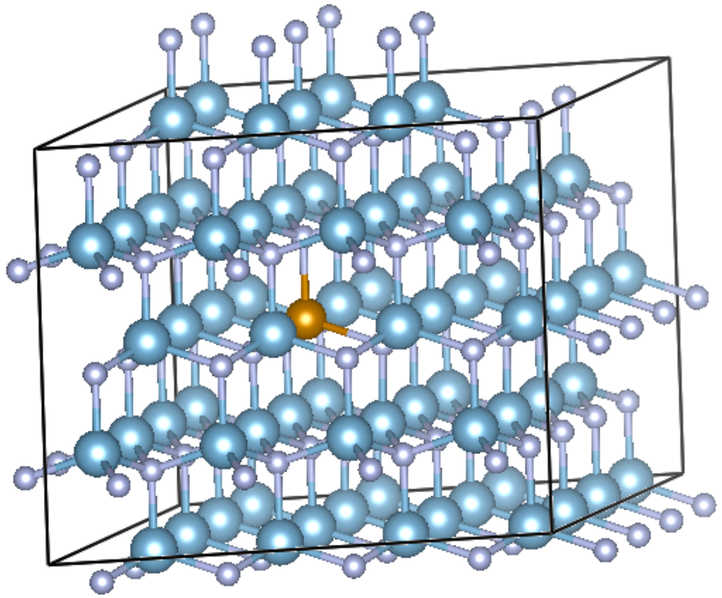
Downfolding interacting models in the constrained random phase approximation (cRPA)

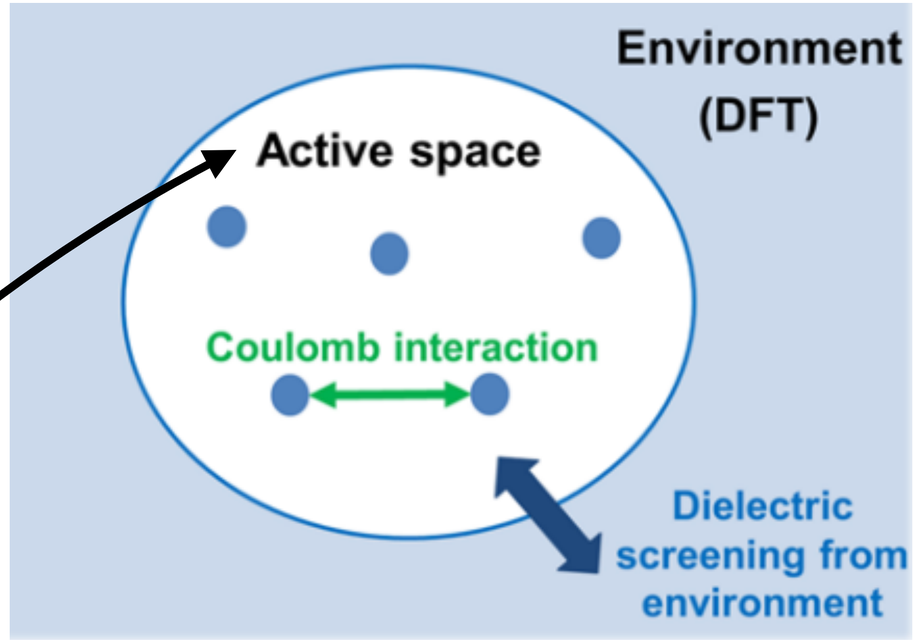
Demonstrating with an Fe impurity in an AlN crystal



Kevin Kleiner
Downfolding School April 29, 2022

Describe an active space and environment

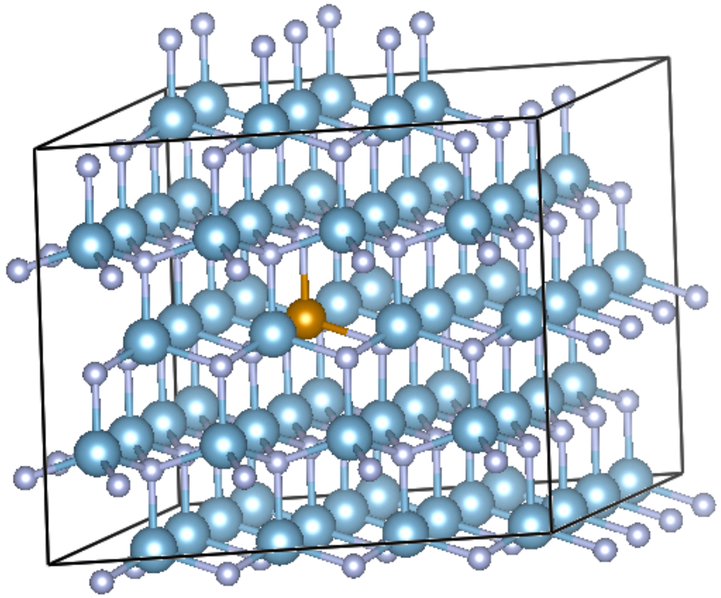
Consider the situation in the cartoon below

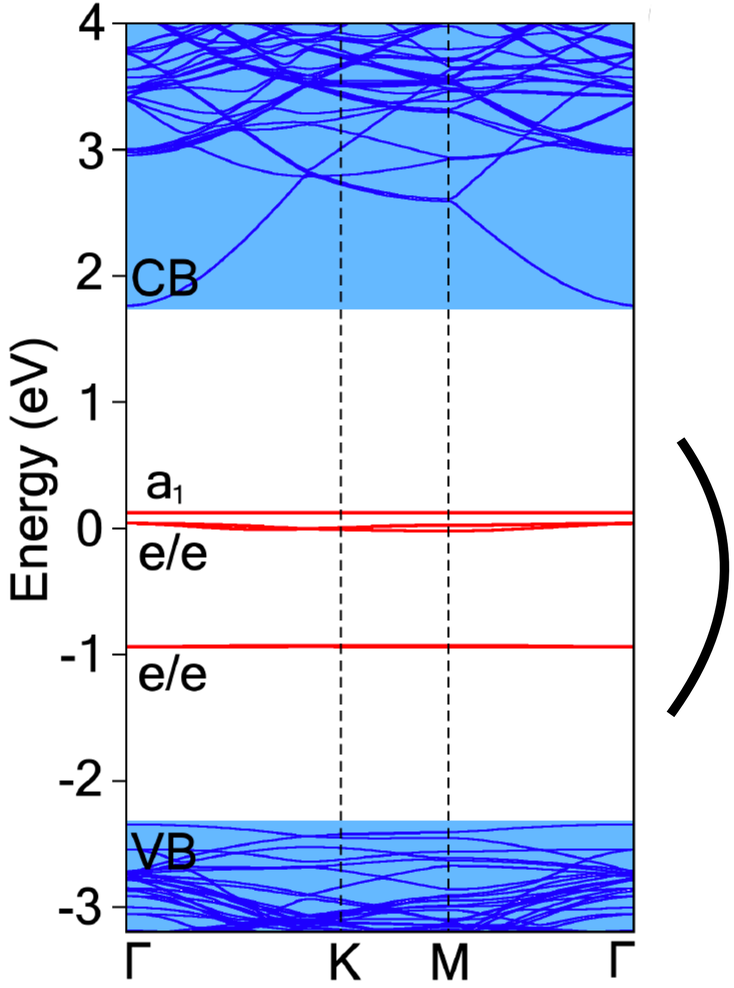


$$\hat{H}_{eff} = -\sum_{ij,\sigma \in Active} (t_{ij}\hat{c}^{\dagger}_{i,\sigma}\hat{c}_{j,\sigma} + hc) + \frac{1}{2}\sum_{ijkl,\sigma\sigma' \in Active} U_{ijkl}\hat{c}^{\dagger}_{i,\sigma}\hat{c}^{\dagger}_{j,\sigma}\hat{c}_{k,\sigma'}\hat{c}_{l,\sigma'}$$

Picture from: H. Ma, M. Govoni, and G. Galli, npj Comput. Mater. **6** 85 (2020).

Concrete example: describing excitations of an Fe impurity in AlN*





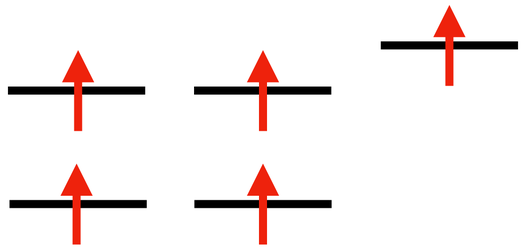
We expect the lowest-energy excitations to **only** involve these bands near E_F

A spin-unpolarized KS-DFT calculation shows five in-gap bands with *Fe* 3*d* character

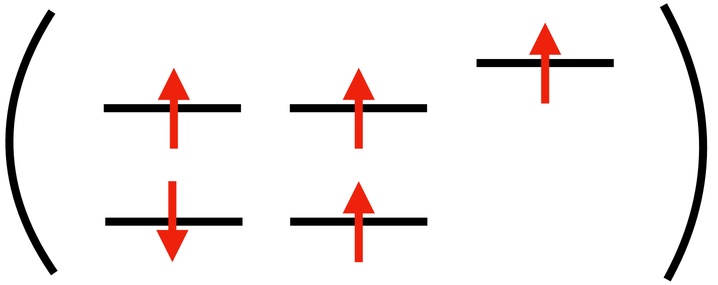
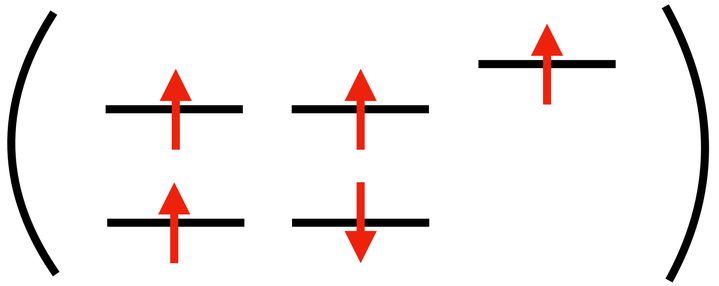
*L. Muechler, et. al., arXiv:2105.08705 (2022).

Fe-in-*AlN* excited states require multi-determinant descriptions

While the spin-5/2 ground state* can be described by singly-occupying five *Fe* 3*d*-like states



the low-spin excited states require <u>expansions of 5+ determinants</u> to satisfy spin symmetry

c_0  + c_1  + ...

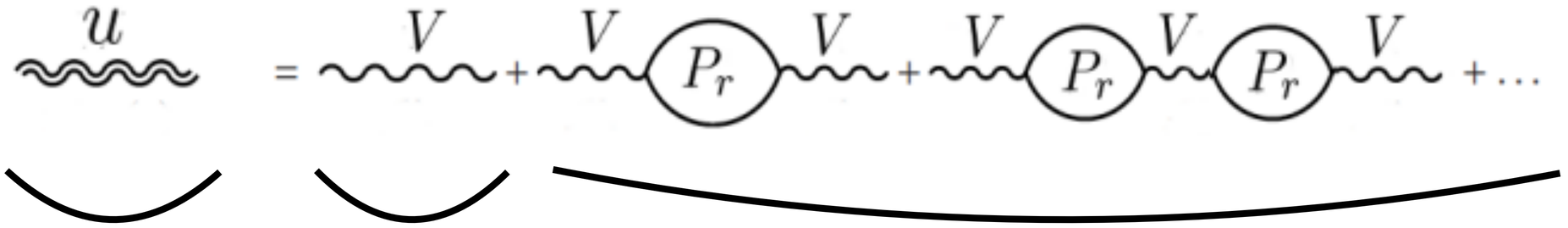
We can describe these low-lying states with an interacting model in the *Fe* 3*d* subspace

*J. Baur, et. al., Appl. Phys. Lett. **65**, 2211 (1994).

Bare coulomb interactions wouldn't be accurate

We need a way to renormalize the 2-body interaction terms in the $Fe\ 3d$ subspace to account for contact with the environment.

Common approach: treat fluctuations in the environment as a perturbation to the bare interaction $\hat{V}$



Screened interaction

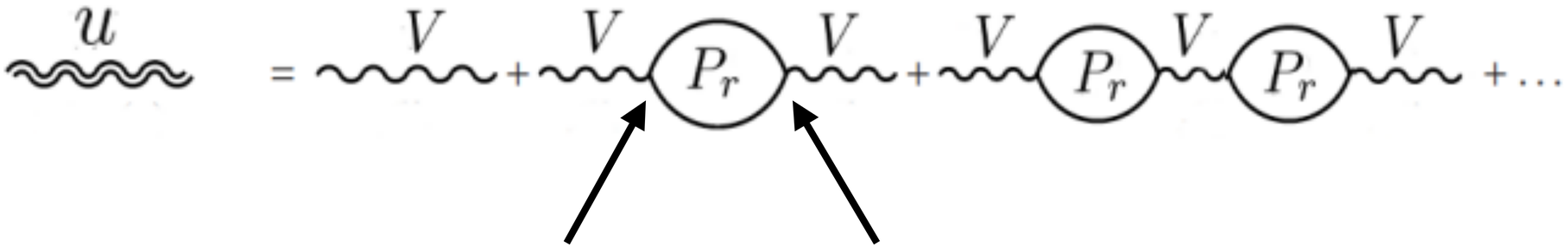
Bare interaction

Correction terms (to be discussed)

Estimate the screened interaction by summing over diagrams

Obtaining the exact sum is intractable, so we truncate the perturbation series with cRPA

Assumption: the only significant corrections come from bubble diagrams



Creation of a virtual particle-hole pair

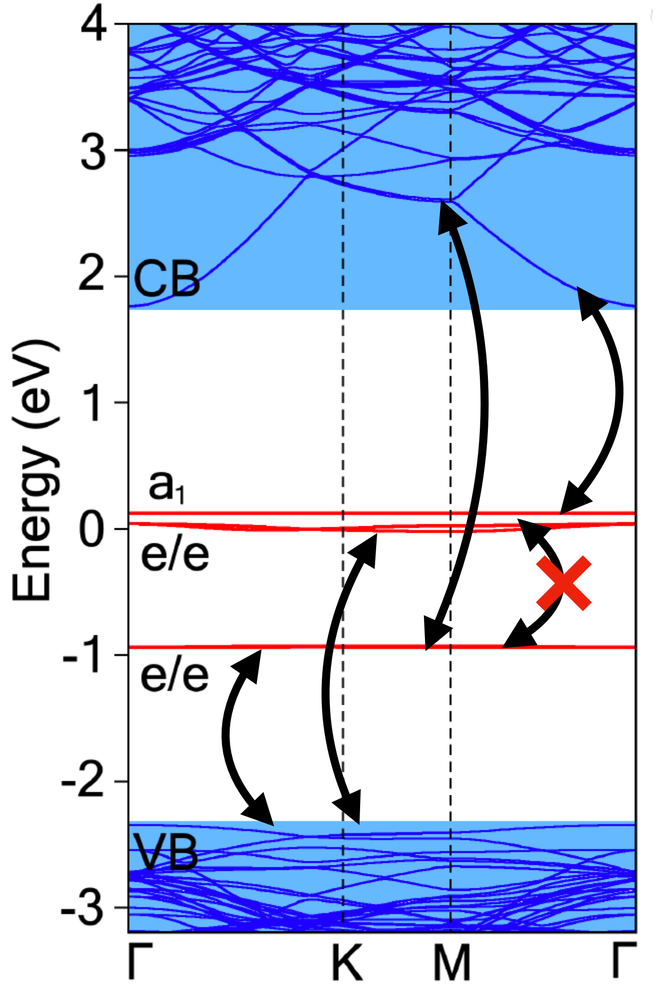
Annihilation of a virtual particle-hole pair

Simplified series sums to $U(\mathrm{r},\mathrm{r}';\omega) = \left[1 - V(\mathrm{r},\mathrm{r}')P_r(\mathrm{r},\mathrm{r}';\omega)\right]^{-1} V(\mathrm{r},\mathrm{r}')$

*Q. Han, B. Chakrabarti, and K. Haule, arXiv:1810.06116 (2018).

Compute P_r, the charge polarization from the environment

Full polarization is the sum over all particle-hole transitions



$$P_r(\mathrm{r},\mathrm{r}';\omega) = P_{full}(\mathrm{r},\mathrm{r}';\omega) - P_{active}(\mathrm{r},\mathrm{r}';\omega)$$

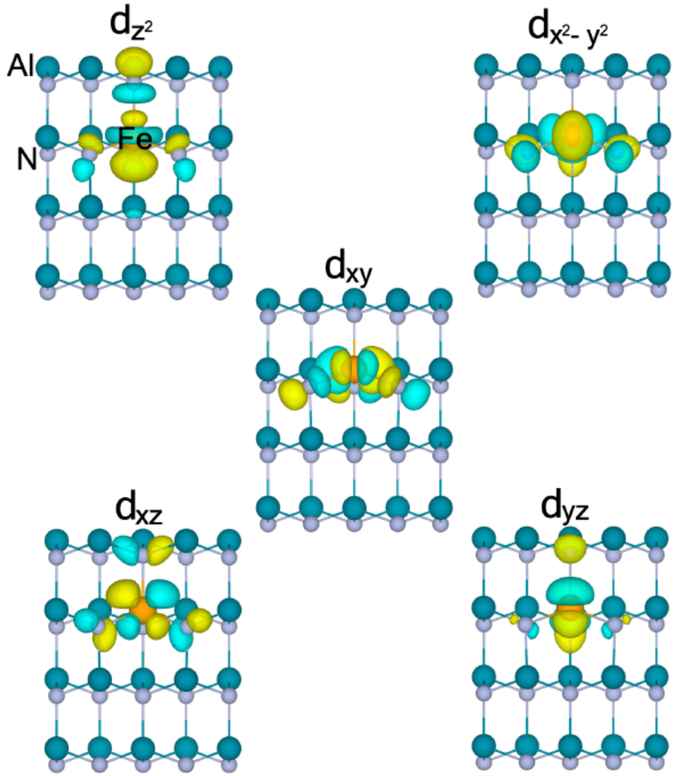
$$P_{full}(\mathrm{r},\mathrm{r}';\omega) = \sum_{n}^{occ}\sum_{n'}^{unocc}\left(\frac{\phi_n(\mathrm{r})\phi_{n'}^*(\mathrm{r})\phi_n^*(\mathrm{r}')\phi_{n'}(\mathrm{r}')}{\omega+(\epsilon_n-\epsilon_{n'})+i\eta}+\frac{\phi_n(\mathrm{r})\phi_{n'}^*(\mathrm{r})\phi_n^*(\mathrm{r}')\phi_{n'}(\mathrm{r}')}{\omega+(\epsilon_{n'}-\epsilon_n)-i\eta}\right)$$

Preview for next week: cRPA downfolding with VASP

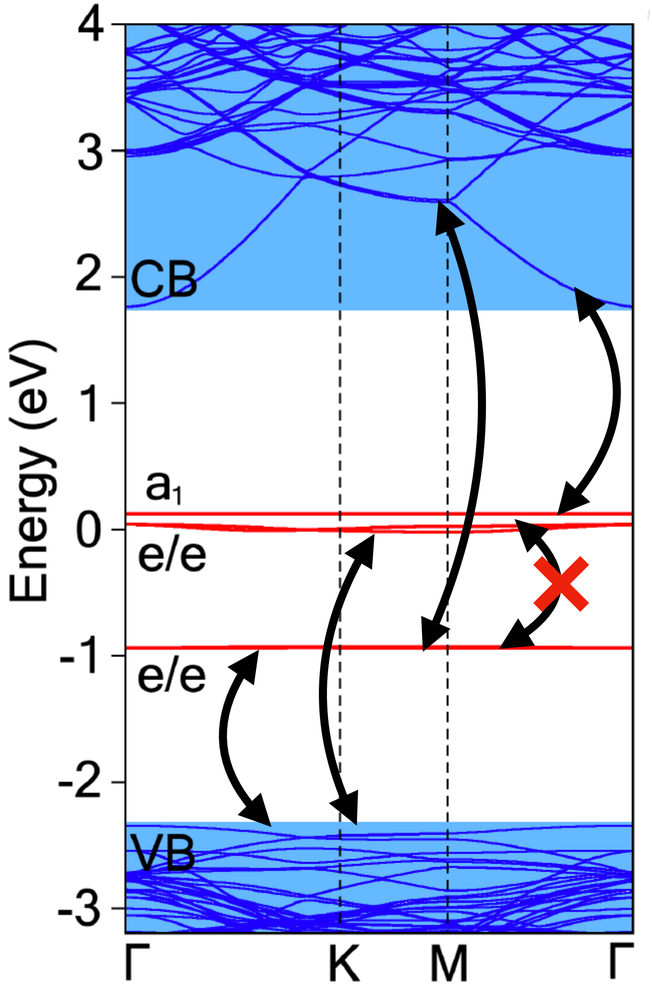
1. Obtain converged $\hat{V}^{KS}$ and $\{\phi_n^{KS}(\mathrm{r})\}$

```
      N        E
DAV:   1     0.217750928225E+04
DAV:   2    -0.971277829749E+03
DAV:   3    -0.110882563137E+04
DAV:   4    -0.110974343615E+04
DAV:   5    -0.110975294626E+04
```

2. Determine the localized 1-body basis and 1-body Hamiltonian



3. Determine the screened interaction and 2-body Hamiltonian



Discussion question: What are the approximations made in each step?