Reduced Density Matrix Functional Theory for Many Electron Systems

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Transition metal oxides–prototypical Mott insulators

Solid	T_N	Band gap
MnO	118	3.7
FeO	198	2.5
CoO	292	2.4
NiO	523	4.1

Table: Néel temperature (in K) and band gap (in eV).

Reduced density matrix

$$\gamma(\mathbf{r},\mathbf{r}') = N \int \Psi(\mathbf{r},\mathbf{r}_2,\ldots,\mathbf{r}_N)\Psi^*(\mathbf{r}',\mathbf{r}_2,\ldots,\mathbf{r}_N)\, d^3r_2 \ldots d^3r_N.$$

Total energy

$$E[\gamma] = \int d^3r' d^3r \delta(\mathbf{r}-\mathbf{r}') \left[-\frac{\nabla^2}{2}\right] \gamma(\mathbf{r},\mathbf{r}') + E_{\mathrm{ext}}[\gamma] + E_{\mathrm{H}}[\gamma] + E_{\mathrm{xc}}[\gamma]$$

Exchange-correlation energy functionals

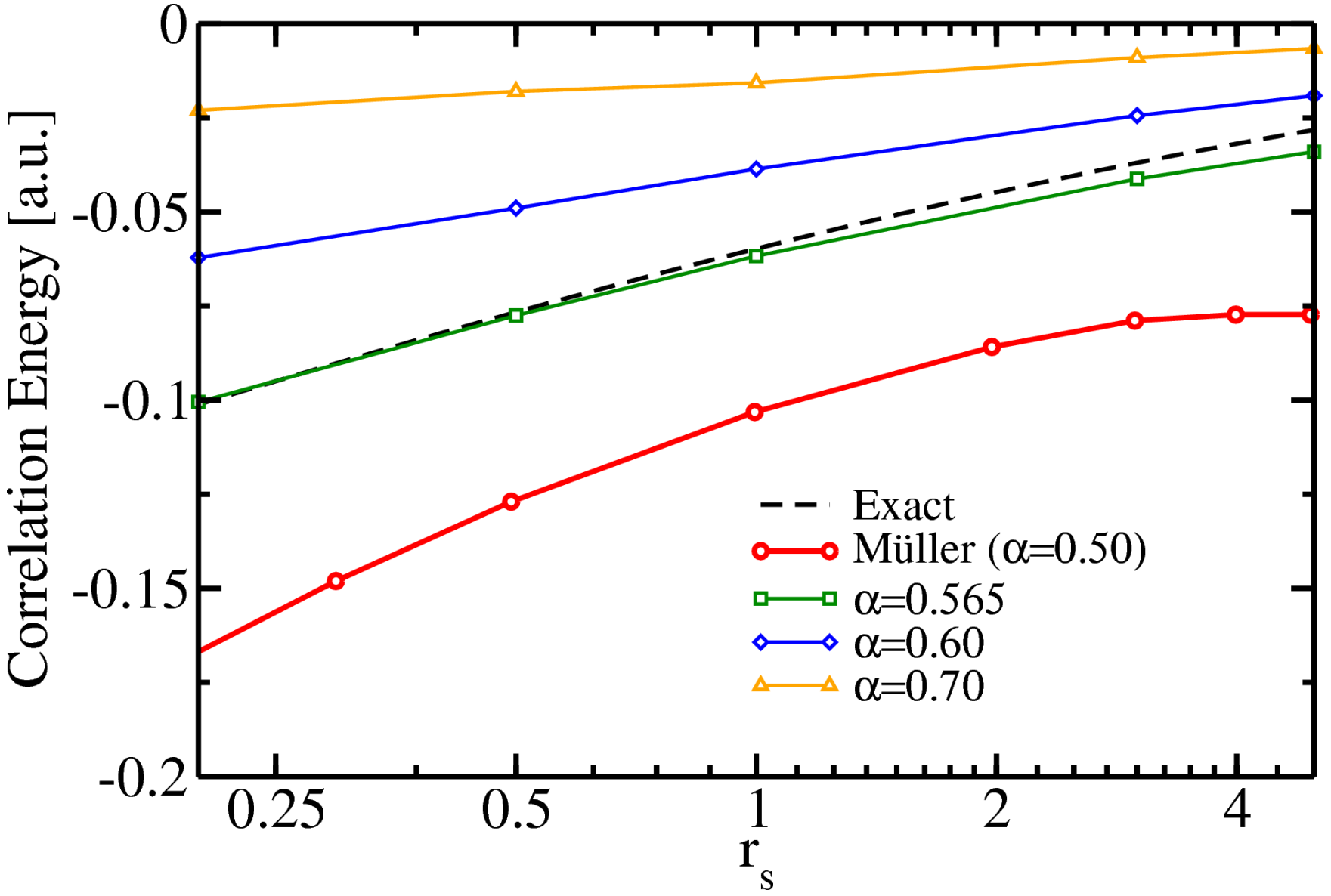
"Power functional", Sharma et al. Phys. Rev. B **78**, R201103 (2008)

RDM energy functional

$$E_{\mathrm{xc}}[\{n_i\},\{\phi_i\}] = -\frac{1}{2}\sum_{ij}(n_i n_j)^{\alpha}\int \frac{\phi_i^*(\mathbf{r})\phi_j(\mathbf{r})\phi_j^*(\mathbf{r}')\phi_i(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d^3r\,d^3r'$$

$$\phi_i(\mathbf{r}) = \sum_l c_l^i \Psi_l^{KS}(\mathbf{r})$$

Fixing the value of α



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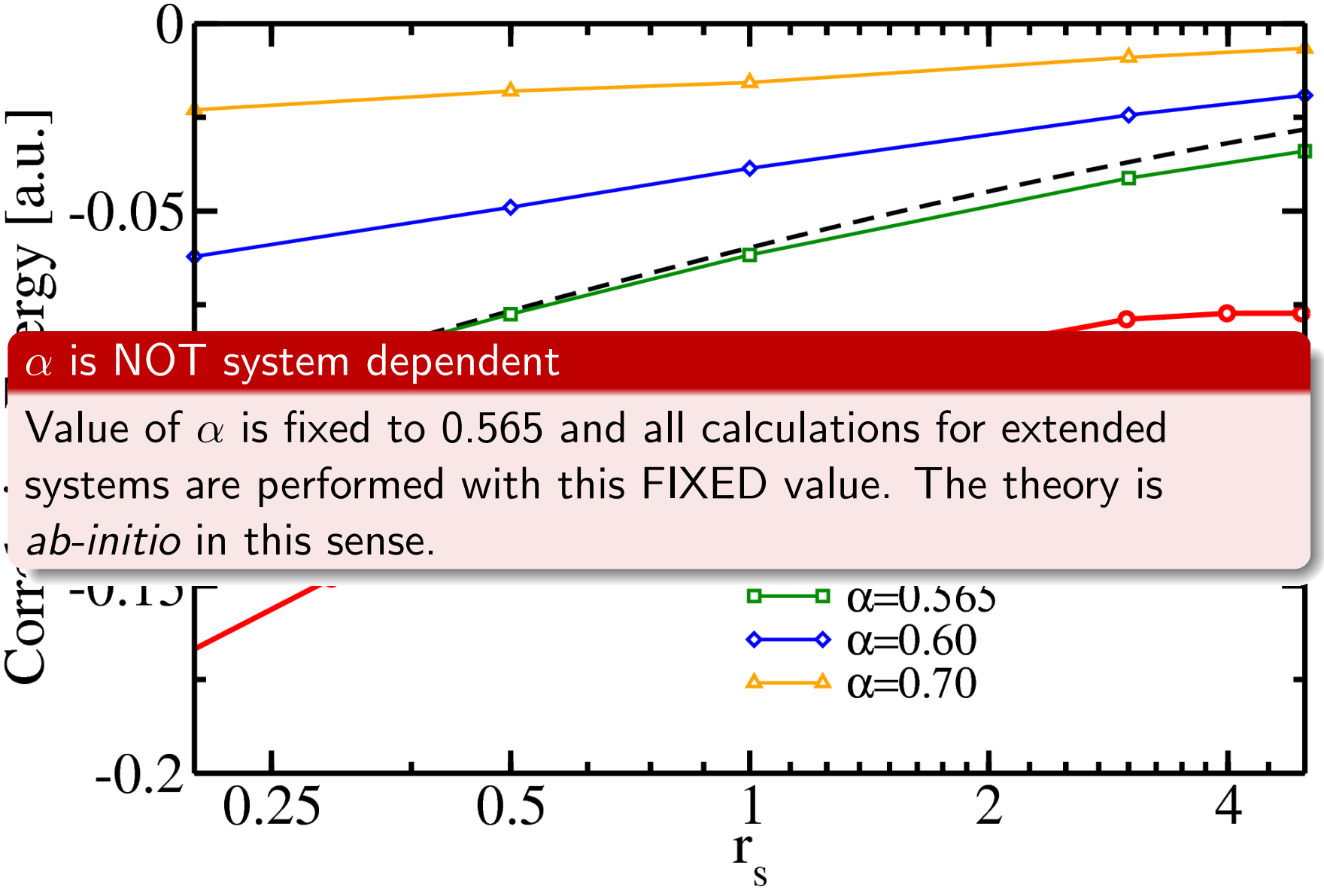

α is NOT system dependent

Value of α is fixed to 0.565 and all calculations for extended systems are performed with this FIXED value. The theory is *ab-initio* in this sense.

Spectral information using RDMFT

Green's function in the bais of natural orbitals:

$$iG_{\mu\beta}(t-t') = \frac{1}{\langle\Psi_0^N|\Psi_0^N\rangle}\langle\Psi_0^N|T[a_\mu(t)a_\beta^\dagger(t')]|\Psi_0^N\rangle$$

Define a restricted but physically significant set of N+1 states

$$\Psi_\zeta^{N+1} = \frac{1}{\sqrt{n_\zeta}}a_\zeta^\dagger|\Psi_0^N\rangle, \quad \Psi_\zeta^{N-1} = \frac{1}{\sqrt{(1-n_\nu)}}a_\zeta|\Psi_0^N\rangle,$$

spectral function

$$A_{\mu\beta}(\omega) = 2\pi\sum_\zeta \frac{1}{n_\zeta}\langle\Psi_0^N|a_\mu a_\zeta^\dagger|\Psi_0^N\rangle\langle\Psi_0^N|a_\zeta a_\beta^\dagger|\Psi_0^N\rangle\delta(\omega-\epsilon_\zeta^+)$$

$$-2\pi\sum_\nu \frac{1}{n_\nu}\langle\Psi_0^N|a_\mu^\dagger a_\nu|\Psi_0^N\rangle\langle\Psi_0^N|a_\nu^\dagger a_\beta|\Psi_0^N\rangle\delta(\omega-\epsilon_\nu^-)$$

$$\mathrm{DOS} = 2\pi\sum_\zeta n_\zeta\delta(\omega-\epsilon_\zeta^+) - 2\pi\sum_\nu(1-n_\nu)\delta(\omega-\epsilon_\nu^-),$$

Spectral information using RDMFT

$$\epsilon_\nu^\pm = E_\nu(N \pm 1) - E(N)$$

with $E_\nu(N \pm 1)$ is energy of periodically repeated Born-von Karman (BvK) cell.

Approximating this

- The only occupation number that will change is the one that corresponds to the very same **k**
- Derivative instead of energy difference

$$\epsilon_\nu^\pm = \left.\frac{\partial E[\{\phi_\nu\},\{n_\nu\}]}{\partial n_\nu}\right|_{n_{\nu=1/2}}$$

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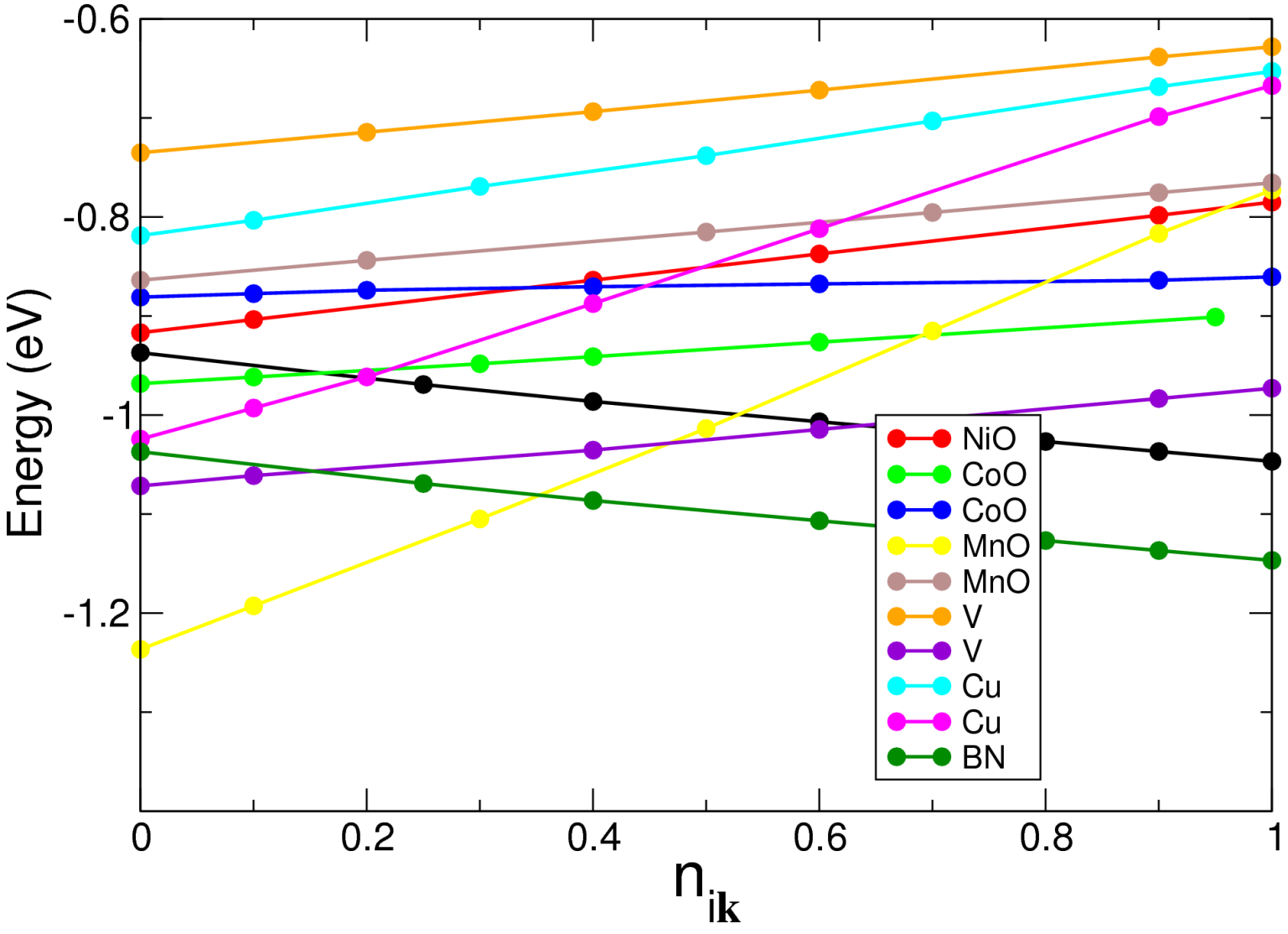

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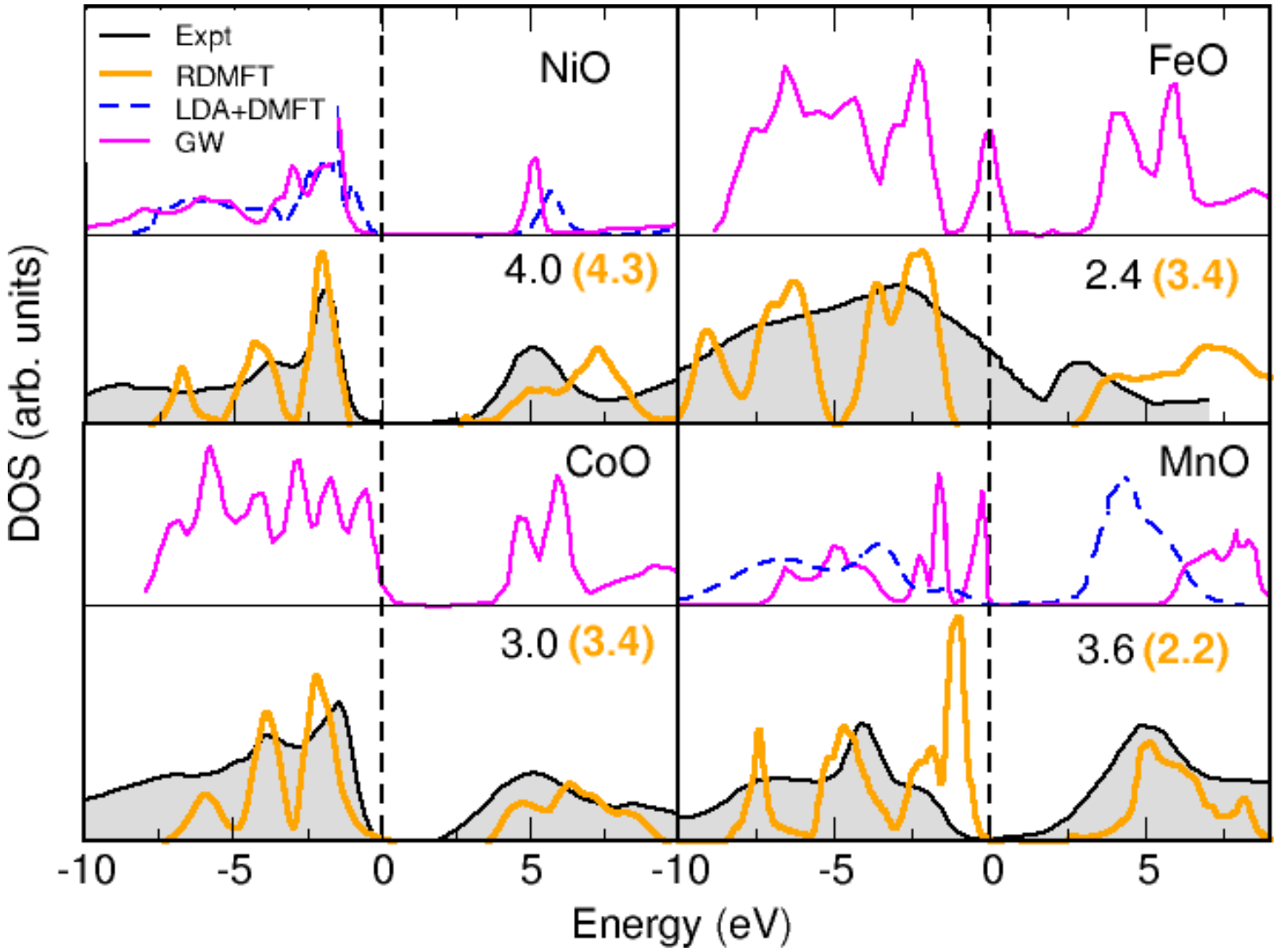
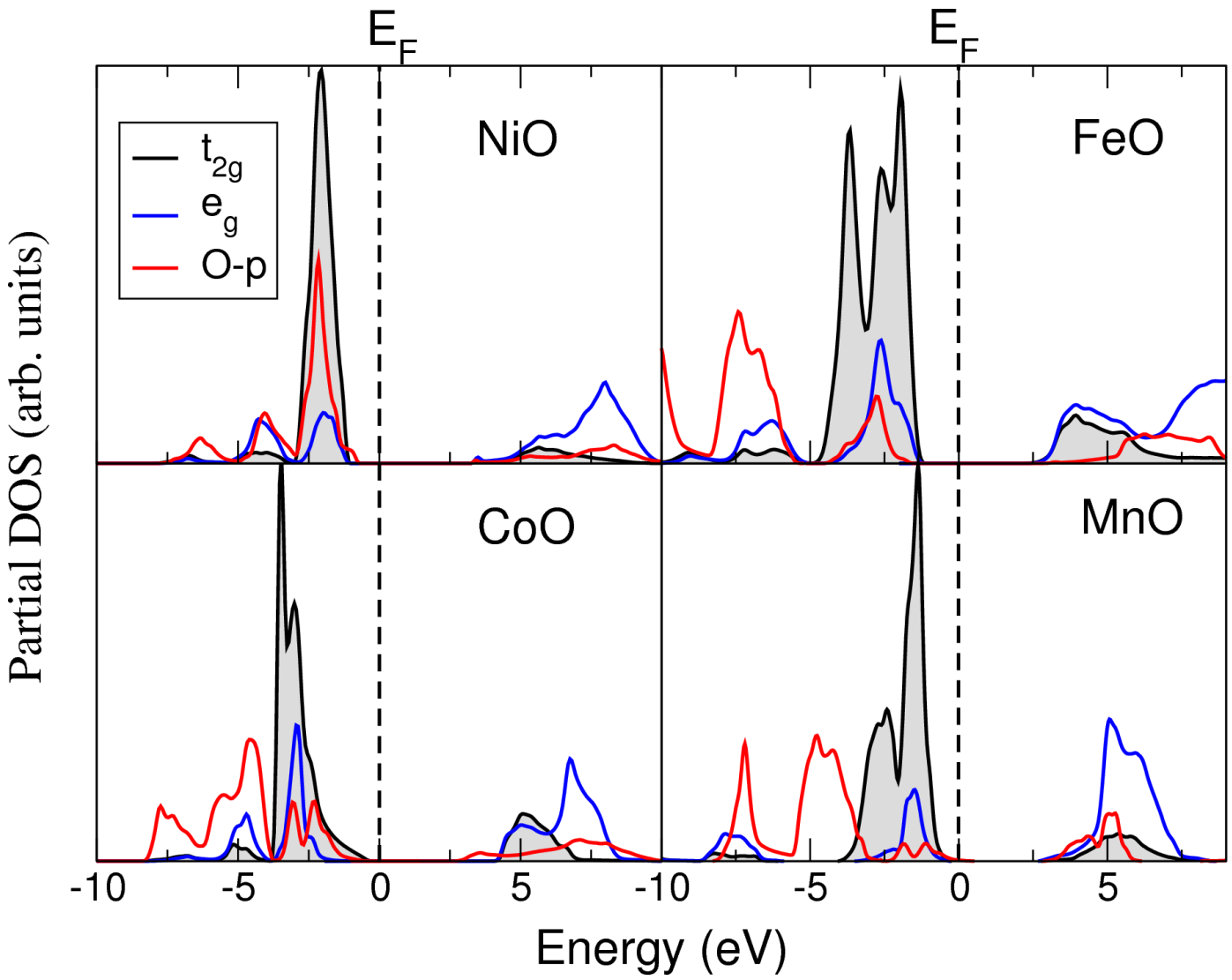
Total DOS for TMOs (without long range magnetic order)



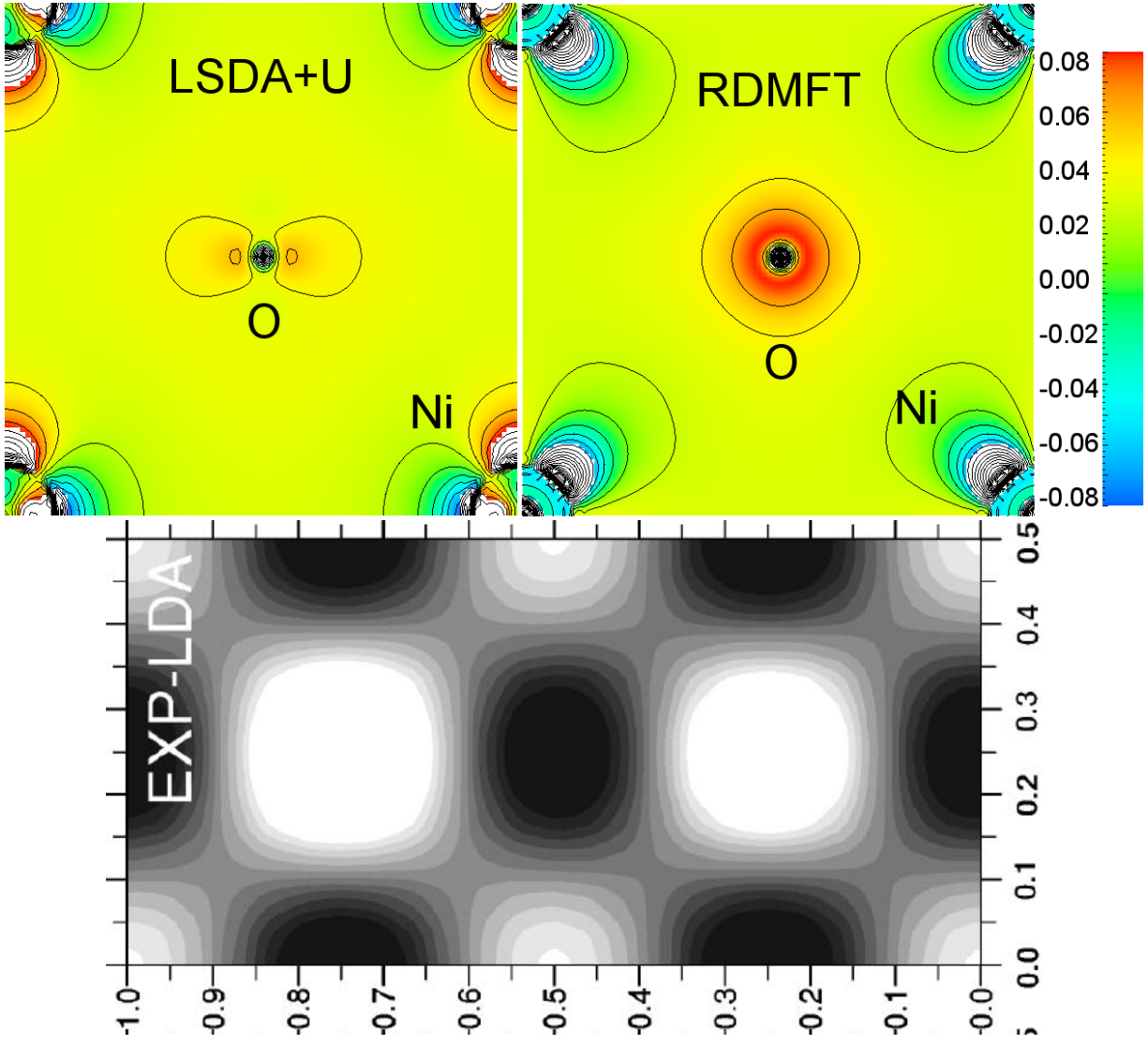
[GW: PRB 79 235114 (09), DMFT: PRB 77 195124 (08), PRB 74 195115 (06), Mat. Mat. 7, 198 (06)]

Partial DOS for TMOs (charge transfer effects)



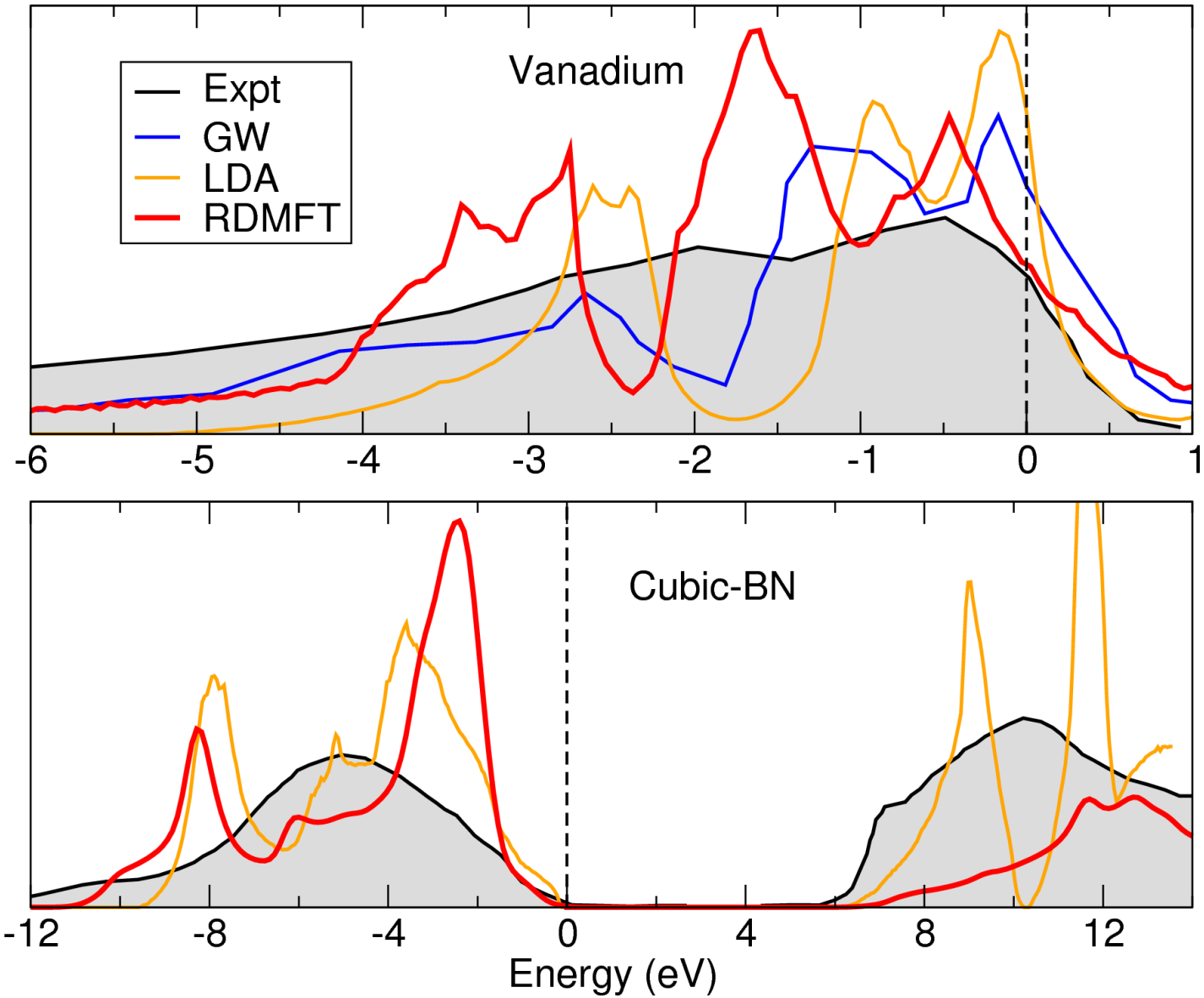
[cond-mat/0912.1118]

Charge density difference for NiO in [110] plane



[cond-mat/0912.1118, Phys. Rev. B **61** 2506 (2000)]

Density of states for metal and band insulator



[cond-mat/0912.1118, PRB 73, 073105 (06)]

Summary and outlook

Summary

- RDMFT has proved to be extremely successful for treatment of finite systems.
- Technique for obtaining spectral information from RDMFT groundstate is proposed.
- RDMFT captures the physics of Mott-insulators.

Outlook

- Magnetic functionals for RDMFT
- Temperature dependent RDMFT to study pahse diagram of TMOs.

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Prize questions

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1. Why is it important the above states are eigen-states
2. When (under what conditions) are these states Eigen-states.