NL-CPA Calculations

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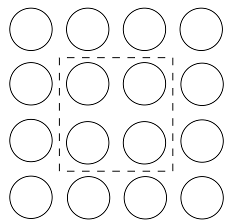
Outline

- Formalism
- Coarse-graining for 3D lattices
- Algorithm
- Results for $CuZn$

Real-Space

Use ECM (Gonis,Stocks,Butler,Winter PRB 29, 555 (1984)). KKR-CPA medium:

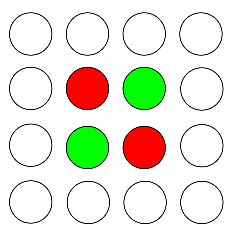
$$\underline{\tau}^{ij} = \underline{t}^{i}\,\delta_{ij} + \sum_{k\neq i} \underline{t}^{i}\,\underline{G}(\mathbf{R}_{ik})\,\underline{\tau}^{kj}$$



$\{I,J\}$

$$\underline{\tau}^{IJ} = \underline{t}_{cl}^{IJ} + \sum_{K,L} \underline{t}_{cl}^{IK}\,\overline{\underline{\Delta}}^{KL}\,\underline{\tau}^{LJ} \quad i.e. \quad \underline{\underline{\tau}}^{-1} = \underline{\underline{t}}_{cl}^{-1} - \underline{\underline{\Delta}}$$

$$where \quad \underline{t}_{cl}^{IJ} = \underline{t}^{I}\delta_{IJ} + \sum_K \underline{t}^{I}\,\underline{G}(\mathbf{R}_{IK})\,\underline{t}_{cl}^{KJ} \quad i.e. \quad \underline{\underline{t}}_{cl}^{-1} = \underline{\underline{t}}^{-1} - \underline{\underline{G}}$$



$\{I,J\}$

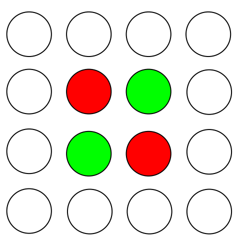
$$\Longrightarrow \underline{\tau}_{imp}^{IJ} = \underline{t}_{cl,imp}^{IJ} + \sum_{K,L} \underline{t}_{cl,imp}^{IK}\,\overline{\underline{\Delta}}^{KL}\,\underline{\tau}_{imp}^{LJ} \quad i.e. \quad \boxed{\underline{\underline{\tau}}_{imp}^{-1} = \underline{\underline{t}}_{cl,imp}^{-1} - \underline{\underline{t}}_{cl}^{-1} + \underline{\underline{\tau}}^{-1}}$$

Averaging over impurity configurations

- Average $\underline{\tau}_{imp}^{IJ}$ over the 2^{N_c} configurations to get $\langle \underline{\tau}_{imp}^{IJ} \rangle$.
- Since $\underline{\underline{\bar{t}}}_{cl}^{-1} = \underline{\underline{\overline{\Delta}}} + \langle \underline{\underline{\tau}}_{imp} \rangle^{-1}$, this generates a new $\underline{\underline{\bar{t}}}_{cl}$ with modified cluster structure constants i.e.

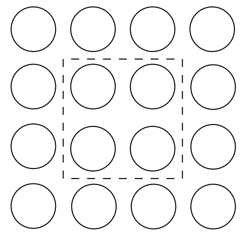
$$
\begin{aligned}
\underline{\underline{\bar{t}}}_{cl}^{-1} &= \underline{\underline{\bar{t}}}^{-1} - \underline{\underline{G}} \\
&\to \underline{\underline{\bar{t}}}^{-1} - \left(\underline{\underline{G}} + \underline{\underline{\delta G}} \right)
\end{aligned}
$$

- Have $\underline{\bar{t}}$ at every site but $\underline{\delta G}(\mathbf{R}_{IJ})$ only between the cluster sites $\{I, J\}$.



- To distribute $\underline{\delta G}(\mathbf{R}_{IJ})$ throughout medium need to develop self-consistent algorithm and treat consistently in reciprocal space.

Reciprocal Space

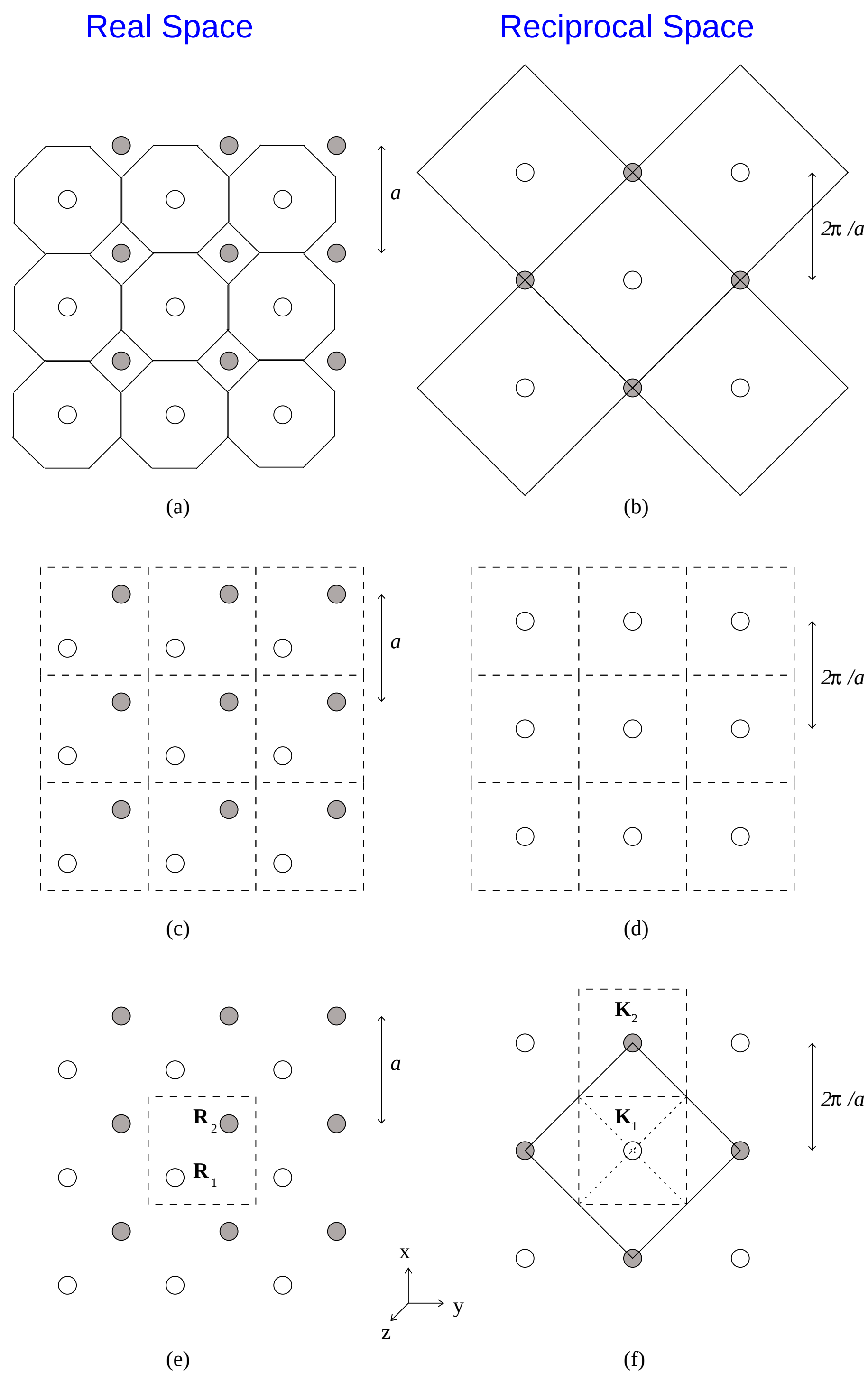


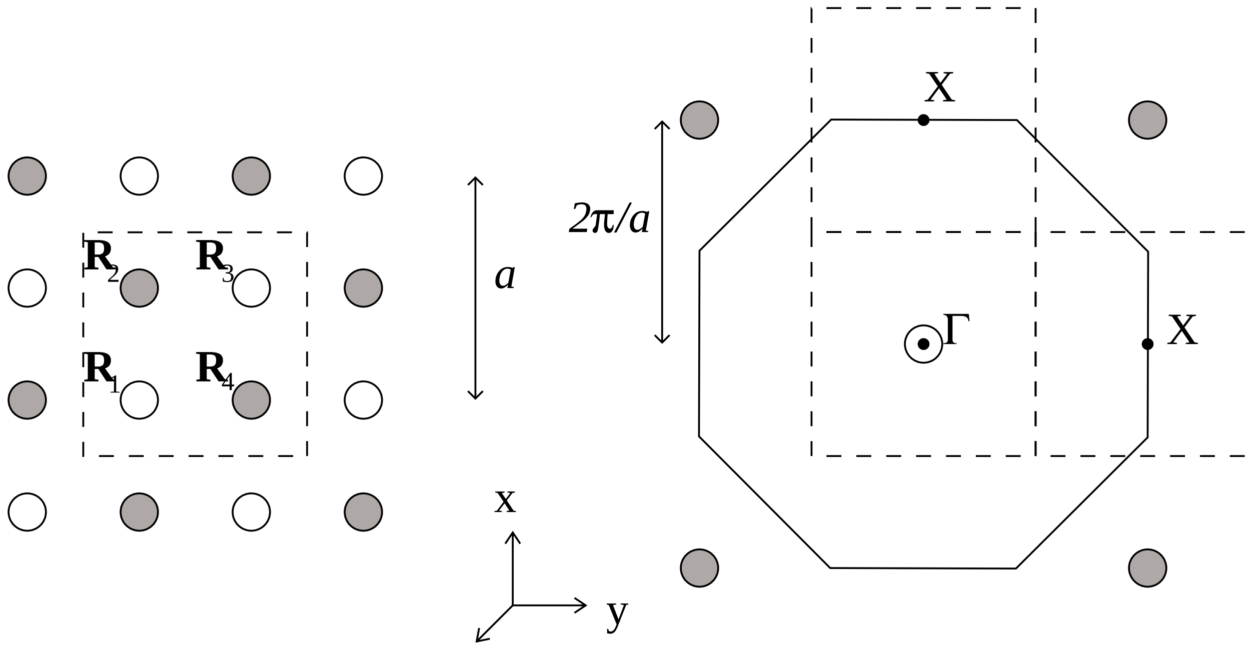
$\{I, J\}$

- $\delta G(\mathbf{R}_{IJ})$ should be a translationally-invariant quantity.
- $\delta G(\mathbf{R}_{IJ})$ should preserve the point-group symmetry of the underlying lattice.
- Enforce these conditions by using coarse-graining idea from DCA
  (M.Hettler *et. al.* PRB **58**, 7475, (1998)), M.Jarrell and H.Krishnamurthy, PRB **63**, 125102, (2001)).
- Approximate $\delta G(\mathbf{k})$ by the set of N_c values $\delta G(\mathbf{K}_n)$ where $n = 1, ..N_c$
- Cluster sites and cluster momenta are related by

$$\frac{1}{N_c} \sum_{\mathbf{K}_n} e^{i\mathbf{K}_n(\mathbf{R}_I - \mathbf{R}_J)} = \delta_{IJ}$$

- First step is to find an appropriate set of cluster sites $\{I, J\}$ and corresponding set of cluster momenta $\{\mathbf{K}_n\}$.


Real Space
Reciprocal Space
a
2π /a
(a)
(b)
a
2π /a
(c)
(d)
a
R2
R1
x
y
z
K2
K1
2π /a
(e)
(f)



(a) (b)

- Using the $IJ \longrightarrow \mathbf{K}_n$ transformation we have

$$\begin{aligned} \underline{\widehat{\delta G}}(\mathbf{R}_{IJ}) &= \frac{1}{N_c} \sum_{\mathbf{K}_n} \underline{\widehat{\delta G}}(\mathbf{K}_n) e^{i\mathbf{K}_n(\mathbf{R}_I - \mathbf{R}_J)} \\ \underline{\widehat{\delta G}}(\mathbf{K}_n) &= \sum_{J \neq I} \underline{\widehat{\delta G}}(\mathbf{R}_{IJ}) e^{-i\mathbf{K}_n(\mathbf{R}_I - \mathbf{R}_J)} \end{aligned}$$

- Can define set of N_c integrals

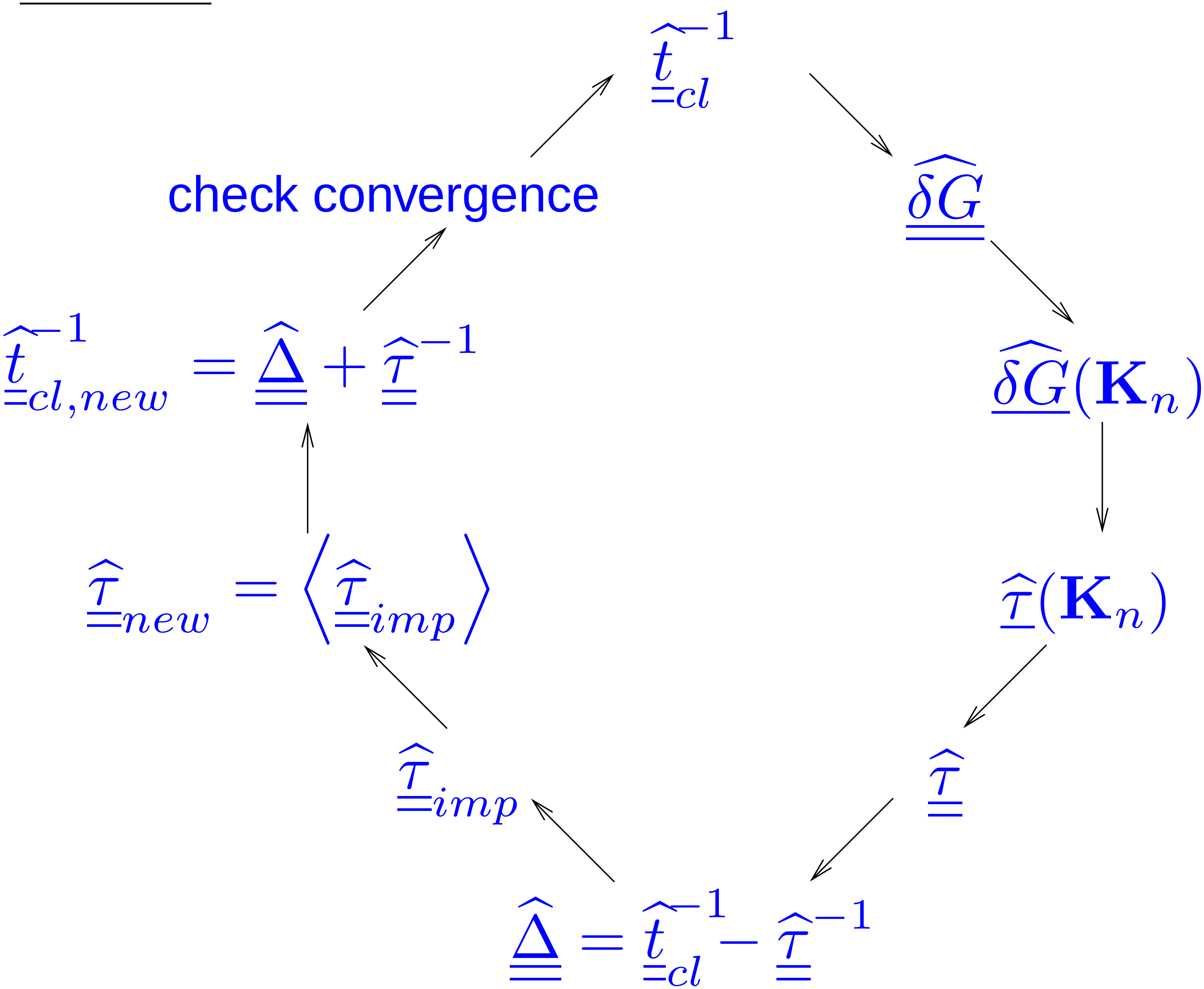
$$\underline{\widehat{\tau}}(\mathbf{K}_n) = \frac{N_c}{\Omega_{BZ}} \int_{\Omega_{\mathbf{K}_n}} d\mathbf{k} \left(\underline{\widehat{t}}^{\,-1} - \underline{G}(\mathbf{k}) - \underline{\widehat{\delta G}}(\mathbf{K}_n) \right)^{-1}$$

- and so

$$\underline{\widehat{\tau}}^{\,IJ} = \frac{1}{\Omega_{BZ}} \sum_{\mathbf{K}_n} \int_{\Omega_{\mathbf{K}_n}} d\mathbf{k} \left(\underline{\widehat{t}}^{\,-1} - \underline{G}(\mathbf{k}) - \underline{\widehat{\delta G}}(\mathbf{K}_n) \right)^{-1} e^{i\mathbf{K}_n(\mathbf{R}_I - \mathbf{R}_J)}$$

- Now possible to contruct self consistent algorithm.

Algorithm


$\underline{\underline{\hat{t}}}_{cl}^{-1}$
$\underline{\underline{\widehat{\delta G}}}$
$\underline{\widehat{\delta G}}(\mathbf{K}_n)$
$\underline{\hat{\tau}}(\mathbf{K}_n)$
$\underline{\underline{\hat{\tau}}}$
$\underline{\underline{\hat{\Delta}}} = \underline{\underline{\hat{t}}}_{cl}^{-1} - \underline{\underline{\hat{\tau}}}^{-1}$
$\underline{\underline{\hat{\tau}}}_{imp}$
$\underline{\underline{\hat{\tau}}}_{new} = \left\langle \underline{\underline{\hat{\tau}}}_{imp} \right\rangle$
$\underline{\underline{\hat{t}}}_{cl,new}^{-1} = \underline{\underline{\hat{\Delta}}} + \underline{\underline{\hat{\tau}}}^{-1}$
check convergence

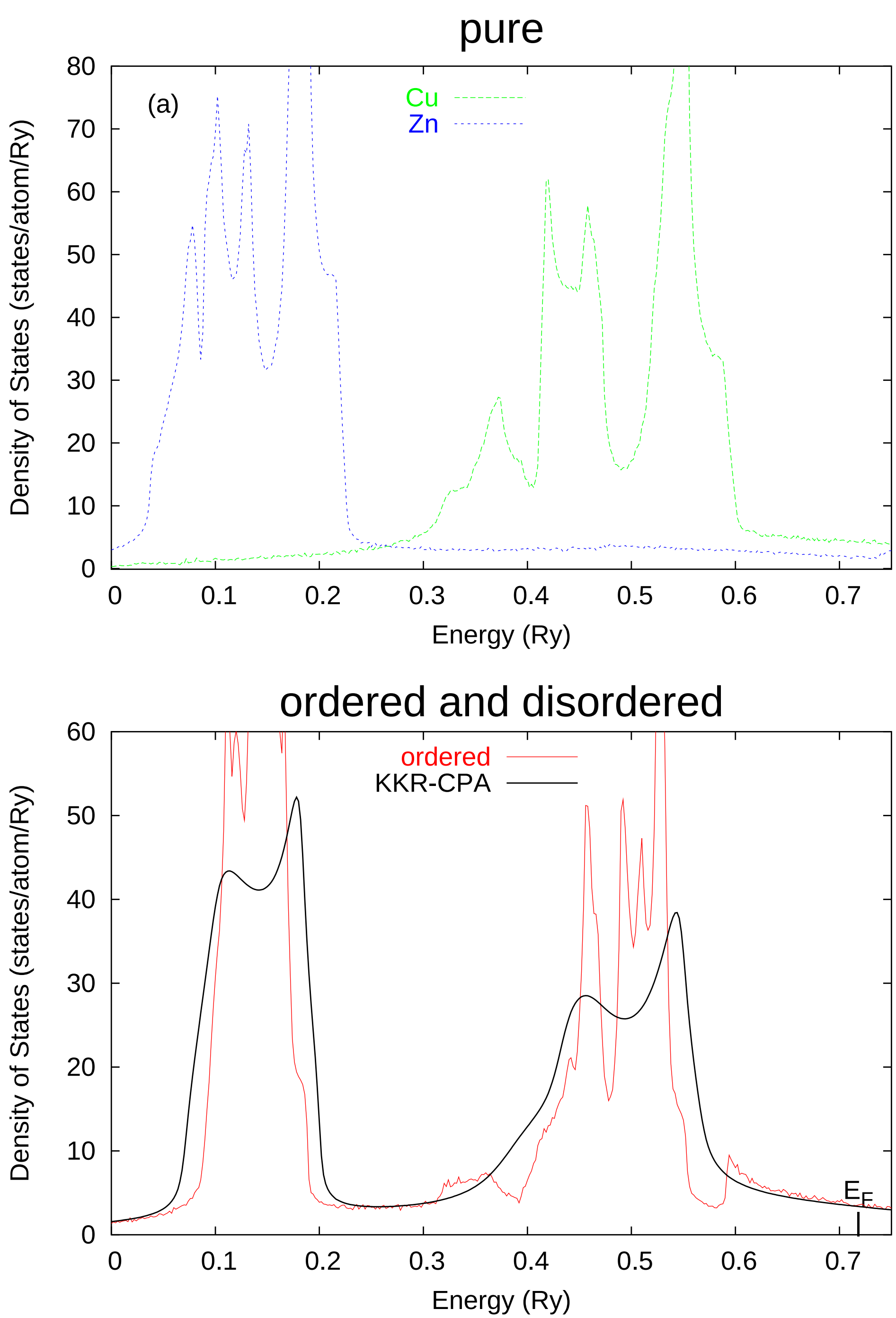
DOS

$$\langle G(E,\mathbf{r}_I,\mathbf{r}'_I)\rangle = \sum_{LL'}\sum_{\gamma}\Big[P(A,\gamma)Z_L^A(E,\mathbf{r}_I)\langle\tau_{LL'}^{II}\rangle_{A,\gamma}\,Z_{L'}^A(E,\mathbf{r}'_I)$$

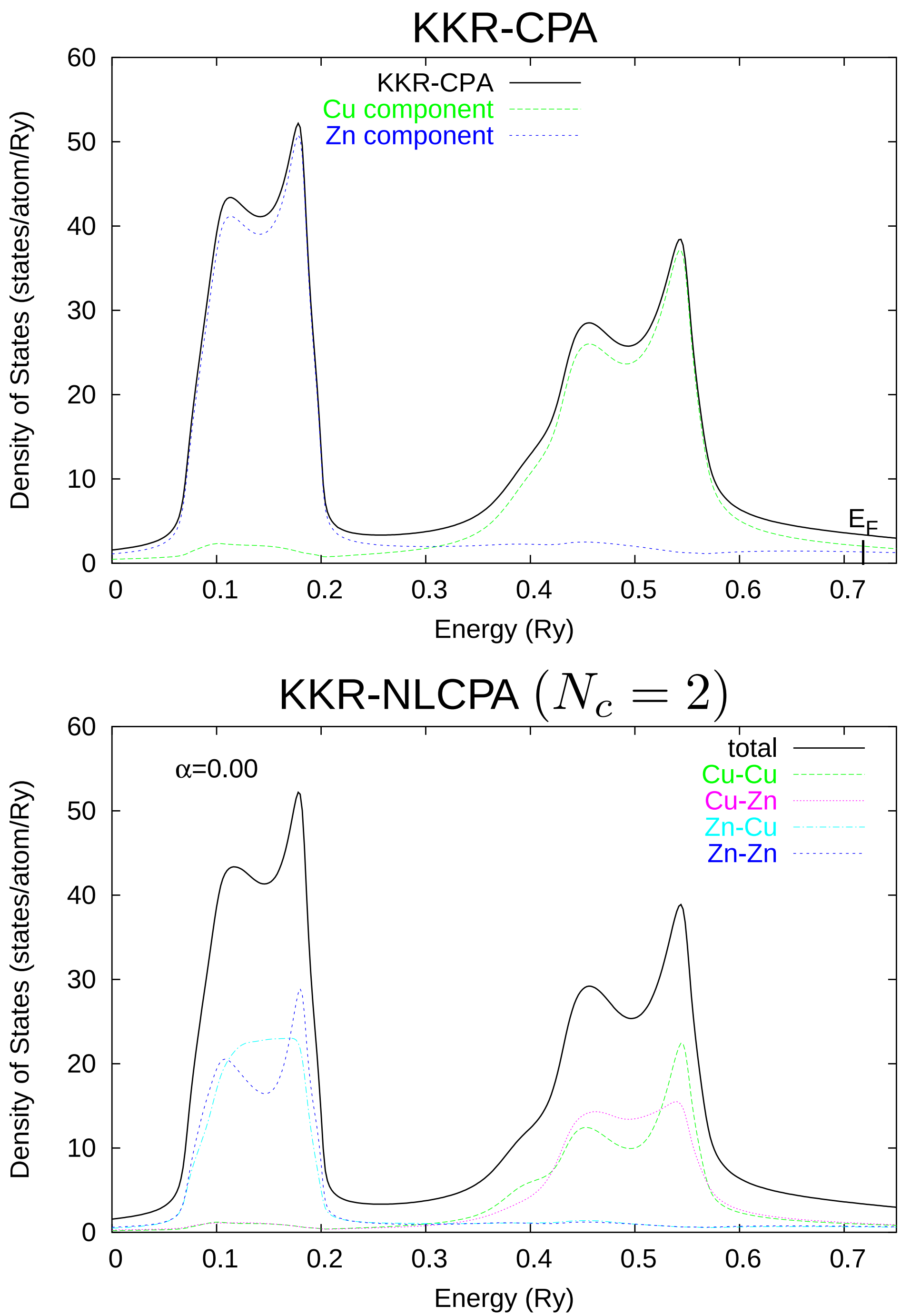
$$+P(B,\gamma)Z_L^B(E,\mathbf{r}_I)\langle\tau_{LL'}^{II}\rangle_{B,\gamma}\,Z_{L'}^B(E,\mathbf{r}'_I)\Big]$$

$$-\sum_L\Big[P(A)Z_L^A(E,\mathbf{r}_I)J_L^A(E,\mathbf{r}'_I)+P(B)Z_L^B(E,\mathbf{r}_I)J_L^B(E,\mathbf{r}'_I)\Big]$$

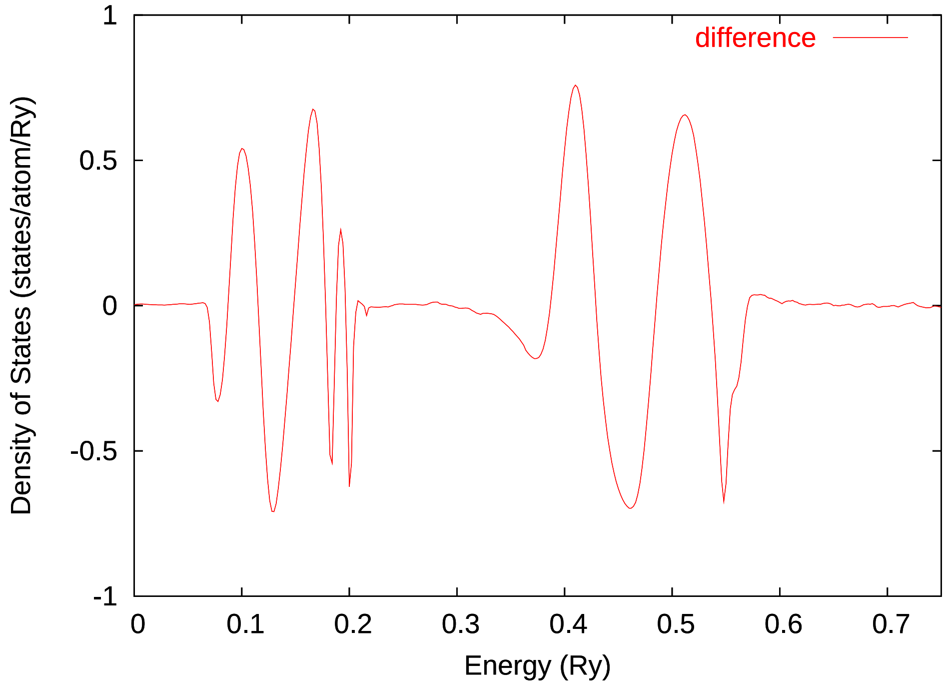
$$n(E) = -\frac{1}{\pi}Im\int_{\Omega_I}\langle G(E,\mathbf{r}_I,\mathbf{r}_I)\rangle d\mathbf{r}_I$$



pure
(a)
Cu
Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
70
80
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
ordered and disordered
ordered
KKR-CPA
E_F
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7



KKR-CPA
KKR-CPA
Cu component
Zn component
Density of States (states/atom/Ry)
Energy (Ry)
E_F
KKR-NLCPA $(N_c = 2)$
α=0.00
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)



difference
Density of States (states/atom/Ry)
Energy (Ry)
1
0.5
0
-0.5
-1
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

Short-Range Order

For $N_c = 2$, introduce nearest neighbour SRO parameter α such that

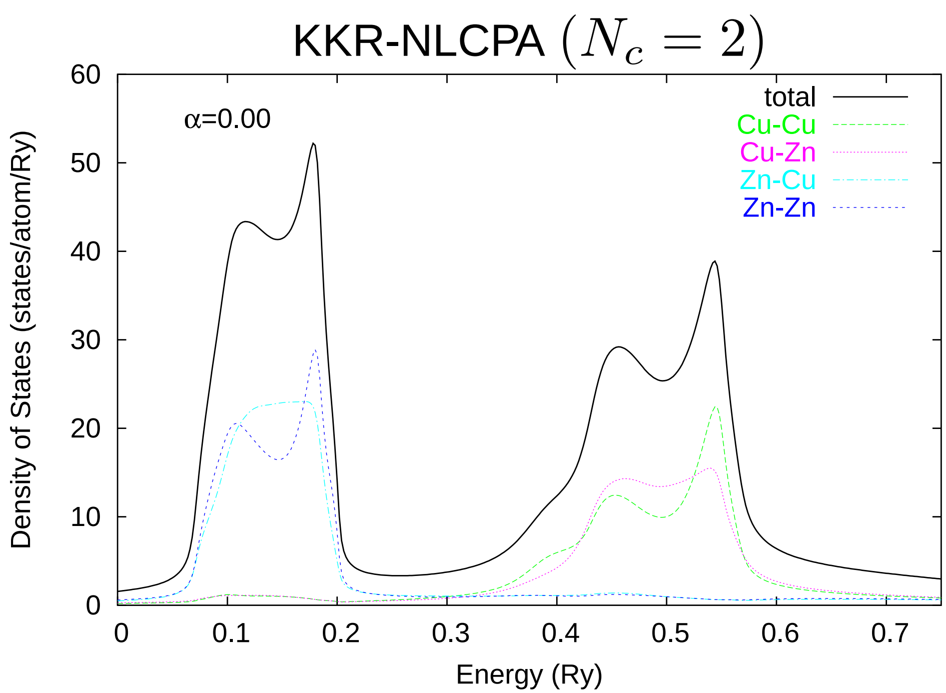
$$P(CuCu) = P(Cu)^2 + \alpha$$
$$P(ZnZn) = P(Zn)^2 + \alpha$$
$$P(CuZn) = P(Cu)P(Zn) - \alpha$$
$$P(ZnCu) = P(Zn)P(Cu) - \alpha$$

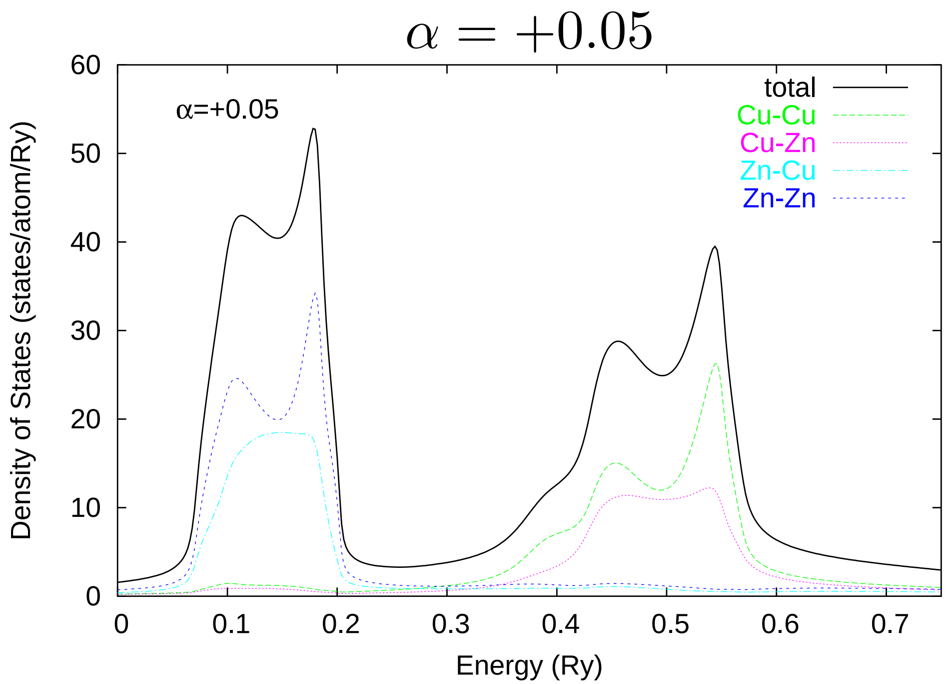
For $Cu_{50}Zn_{50}$, $P(Cu) = P(Zn) = 0.5$ and so

$-0.25 \leq \alpha \leq +0.25$ where

-ve values correspond to short-range ordering

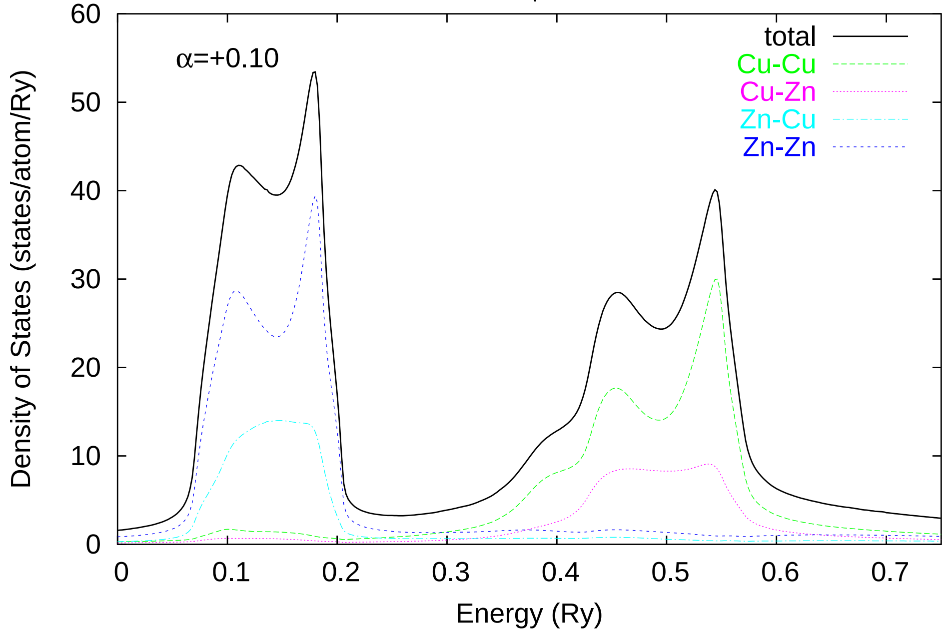
+ve values correspond to short-range clustering


KKR-NLCPA $(N_c = 2)$
α=0.00
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7



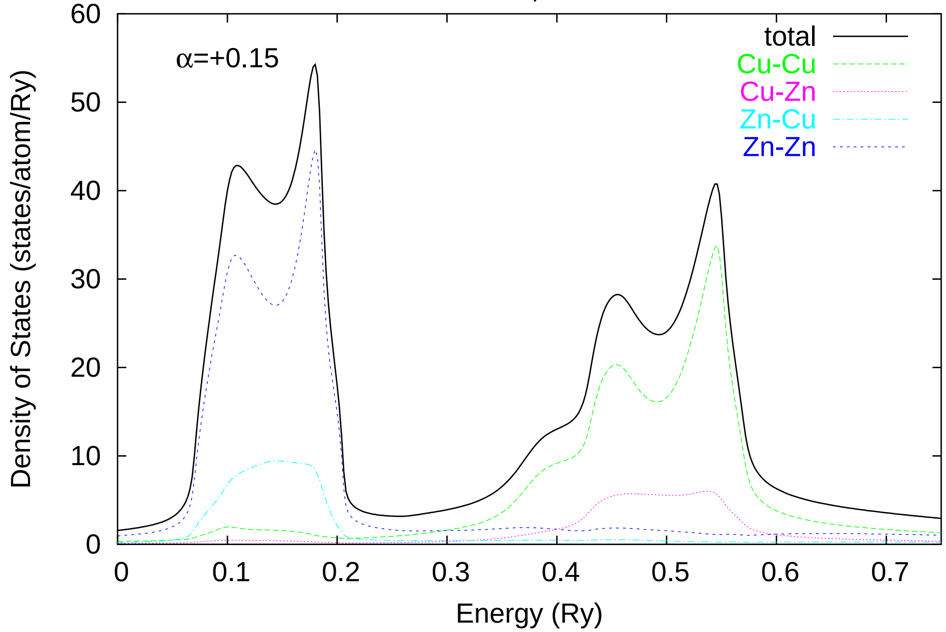
α = +0.05
α=+0.05
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = +0.10$



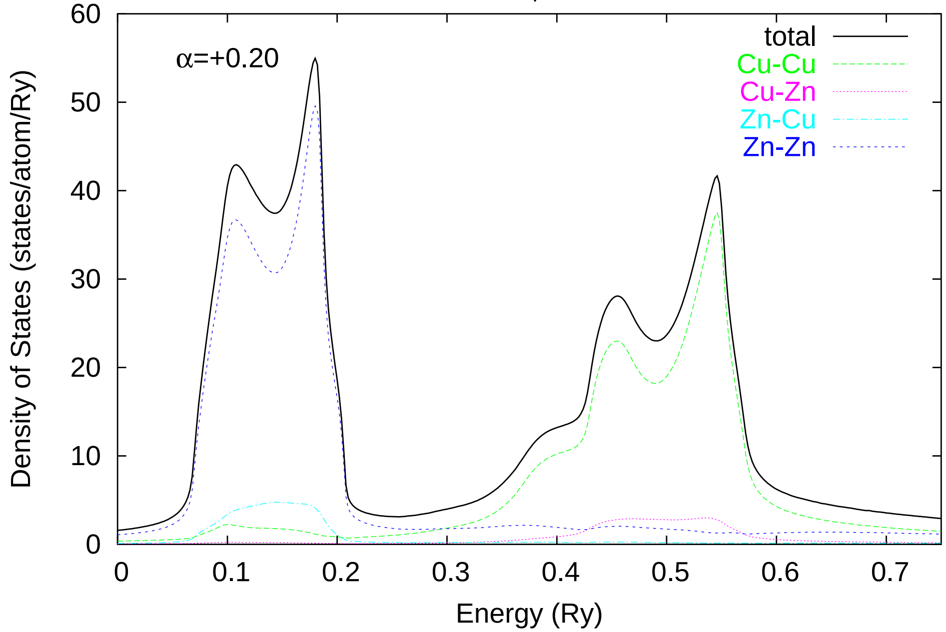
α=+0.10
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = +0.15$

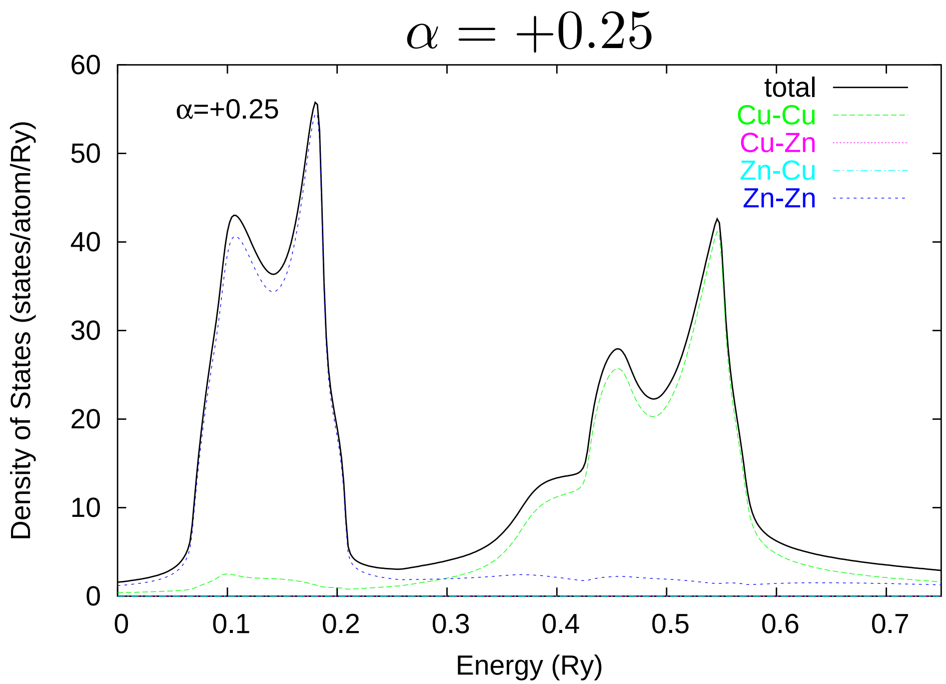


α=+0.15
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = +0.20$

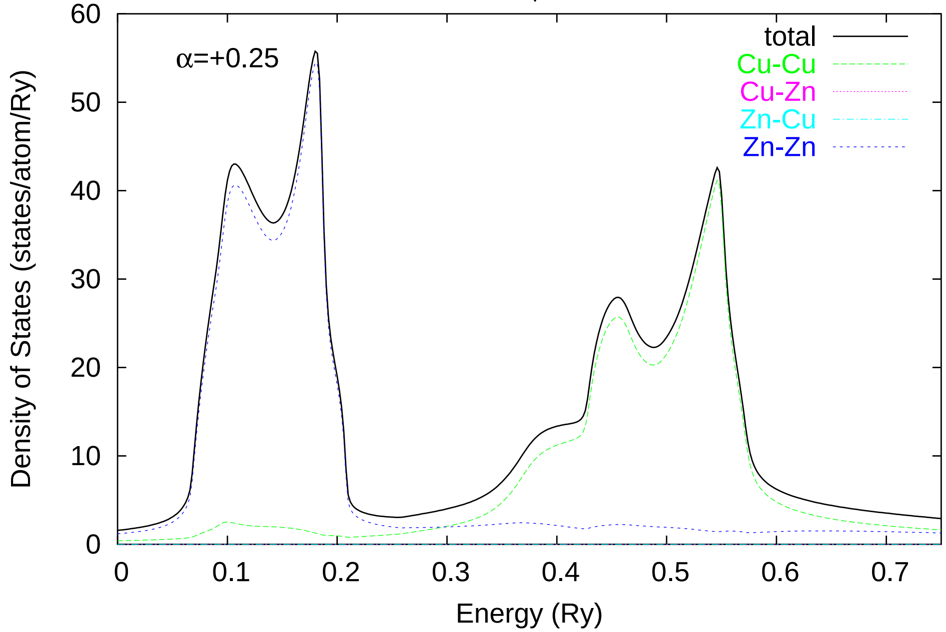


α=+0.20
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0.1
0.2
0.3
0.4
0.5
0.6
0.7

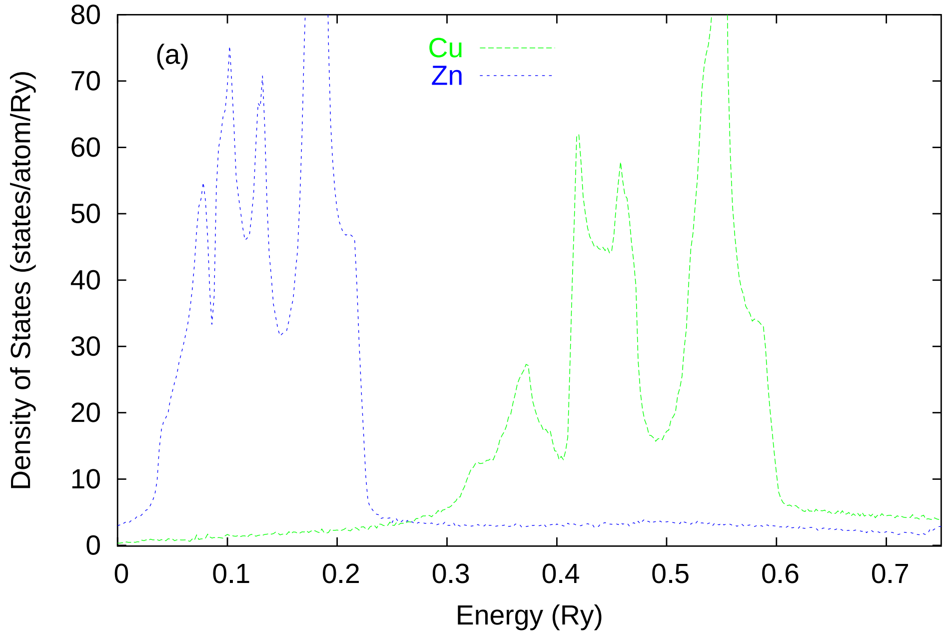


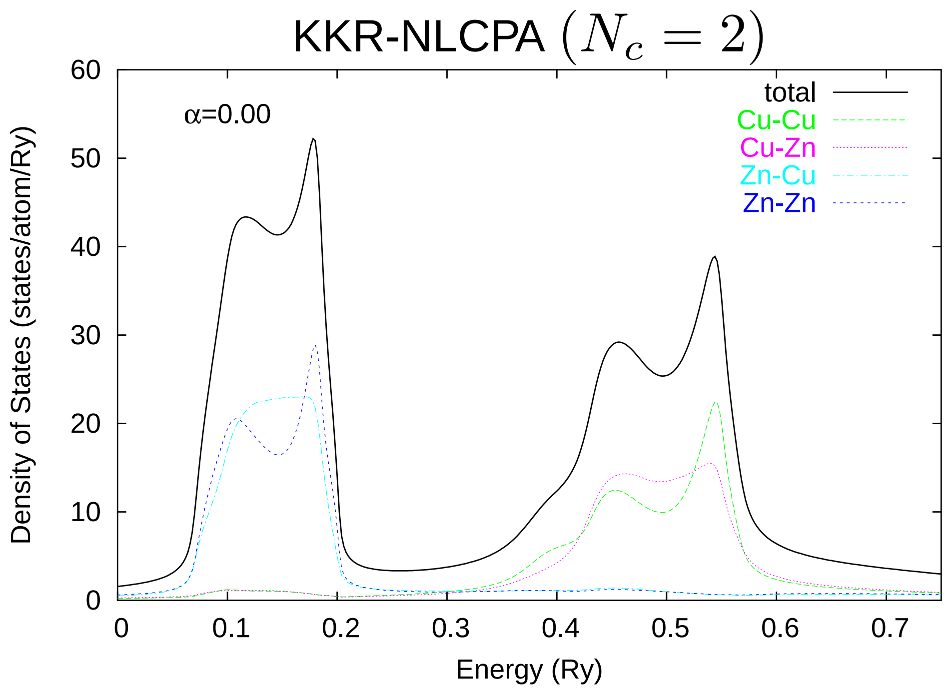
$\alpha = +0.25$
α=+0.25
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = +0.25$



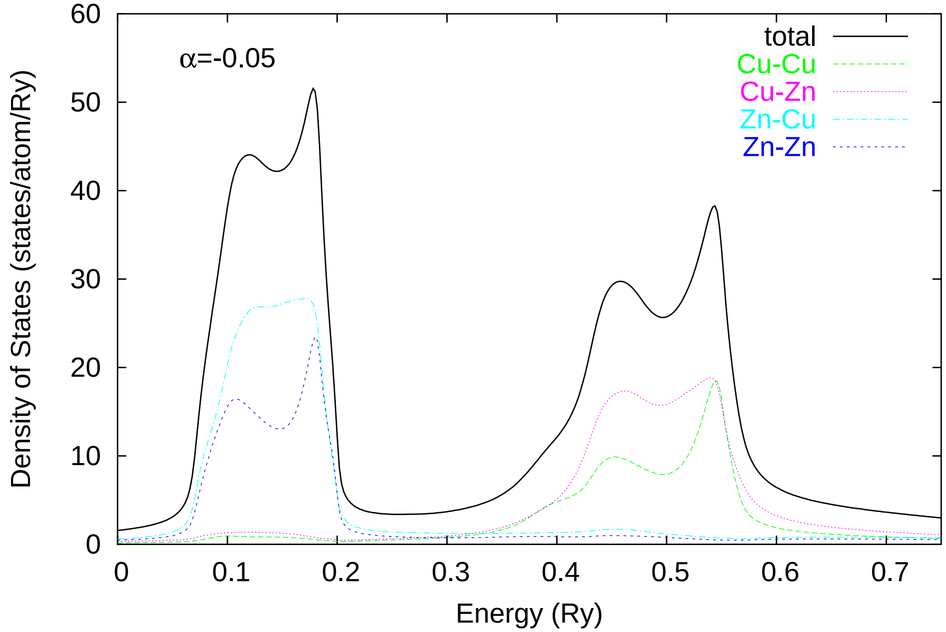
pure





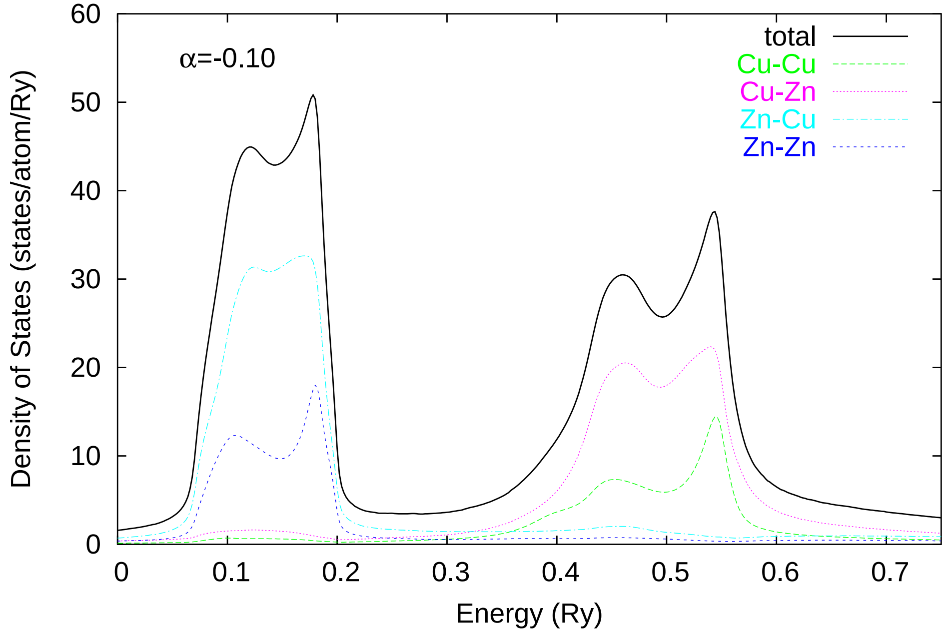
KKR-NLCPA $(N_c = 2)$
α=0.00
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$$\alpha = -0.05$$



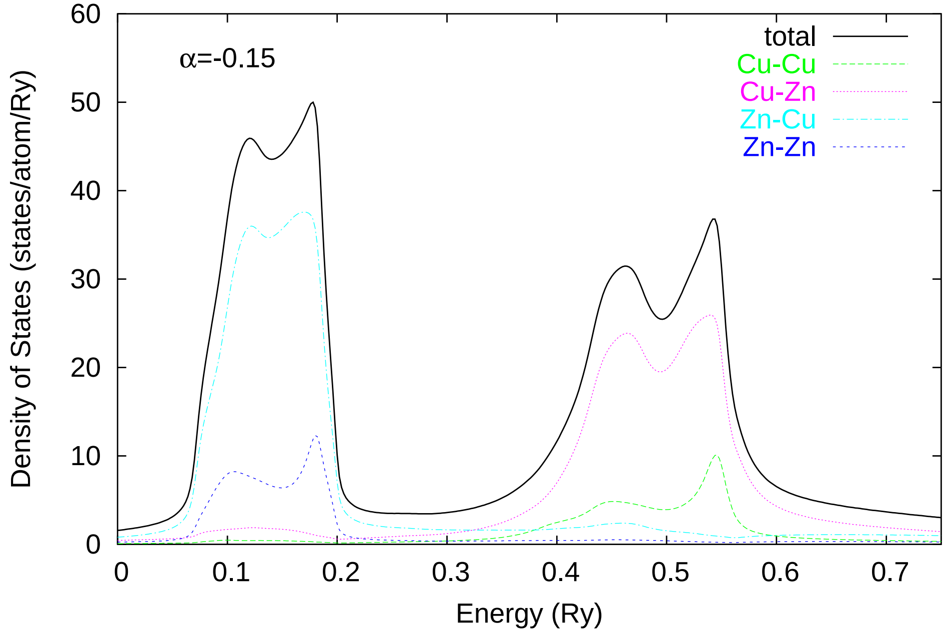
α=-0.05
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = -0.10$



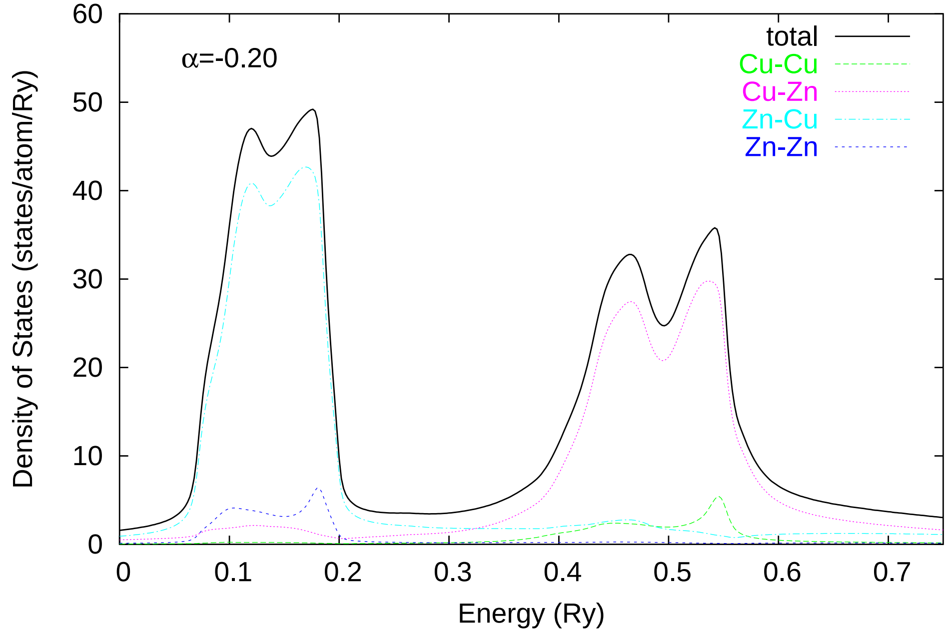
α=-0.10
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = -0.15$



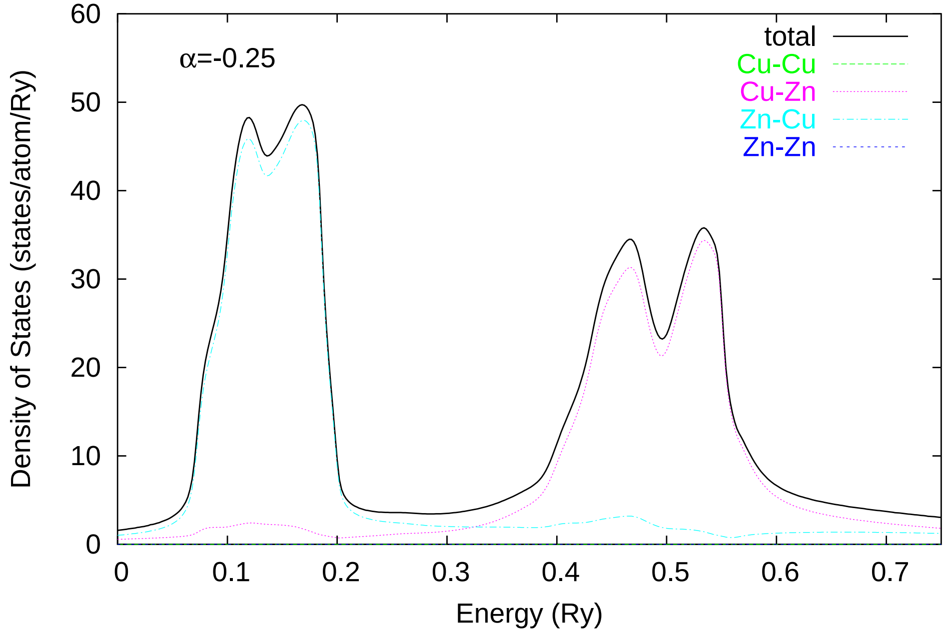
α=-0.15
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = -0.20$



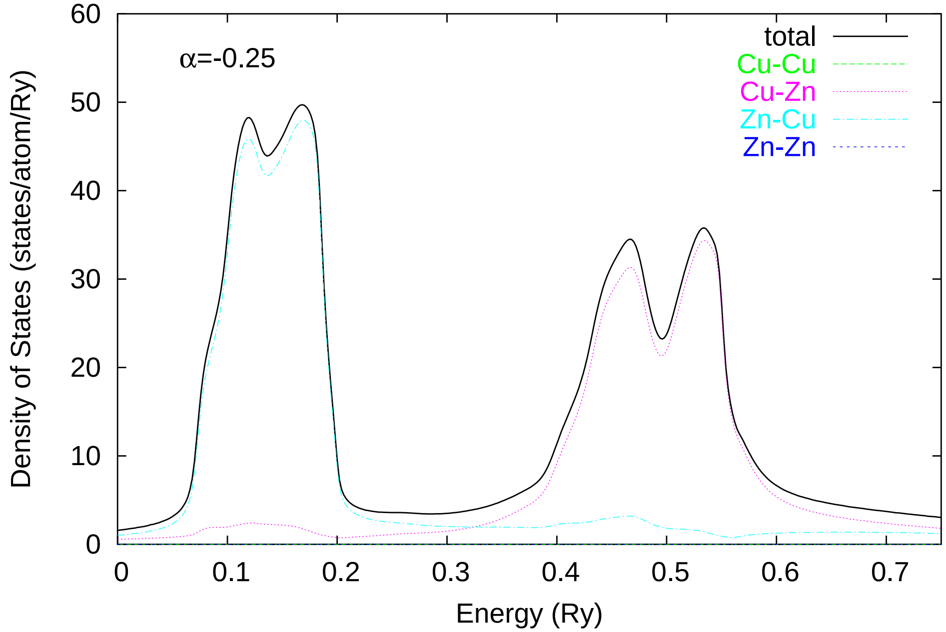
α=-0.20
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$$\alpha = -0.25$$

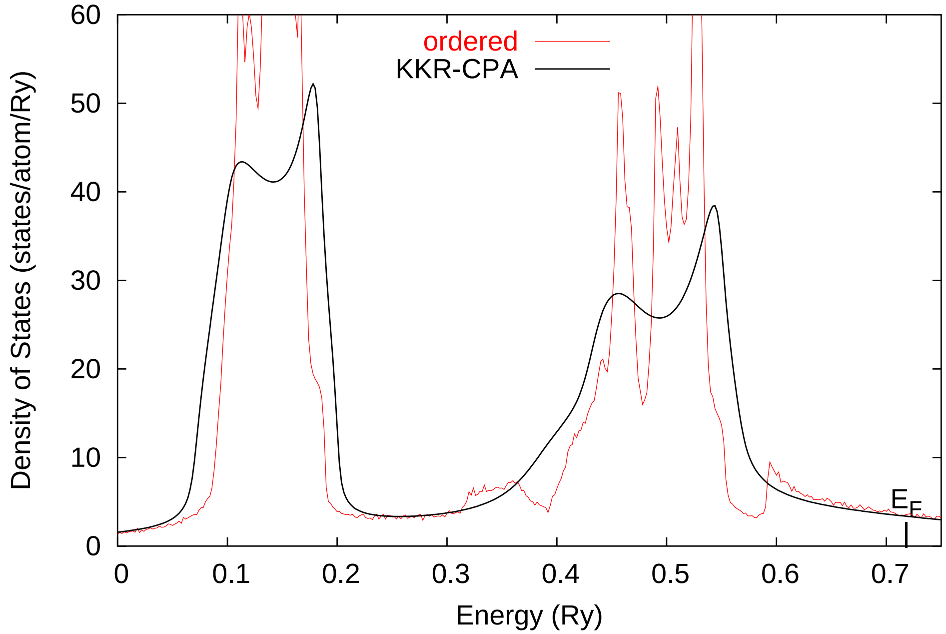


α=-0.25
total
Cu-Cu
Cu-Zn
Zn-Cu
Zn-Zn
Density of States (states/atom/Ry)
Energy (Ry)
0
10
20
30
40
50
60
0.1
0.2
0.3
0.4
0.5
0.6
0.7

$\alpha = -0.25$



ordered and disordered



Papers

Formalism: D.A.Rowlands, J.B.Staunton, B.L.Györffy: PRB 67, 115109 (2003)

3D Results: D.A.Rowlands, J.B.Staunton, B.L.Györffy, E.Bruno, B.Ginatempo: cond-mat/0411347; PRB 70, Issue 20, (May 2005)

DFT: D.A.Rowlands, J.B.Staunton, B.L.Györffy, E.Bruno, B.Ginatempo: to be submitted

Future Work

Include lattice displacements.

Ab-initio theory of SRO: calculate α via linear response and self-consistently feed back into electronic structure.

Acknowledgements

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