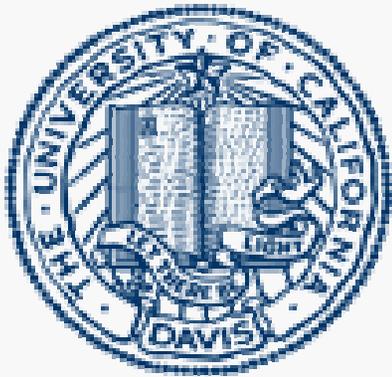


4<sup>th</sup> FPLO Workshop in Dresden

(April 1, 2005)

**CPA approach: On Heavy Carbon**  
**Doping of MgB<sub>2</sub>**



**UC Davis**

**Deepa Kasinathan**

**Kwan-Woo Lee**

**Warren E. Pickett**

**Acknowledgments: P.C. Canfield,  
L.C. Cooley, K. Koepernik, I.I. Mazin,  
and D.J. Singh**

# Outline

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- ◇ Experimental data
- ◇ Computational methods
- ◇ Smeared band structure
- ◇ CPA & Supercell calculation results
- ◇ VCA results
- ◇  $\sigma$  hole concentration
- ◇ Conclusion

# Experimental data

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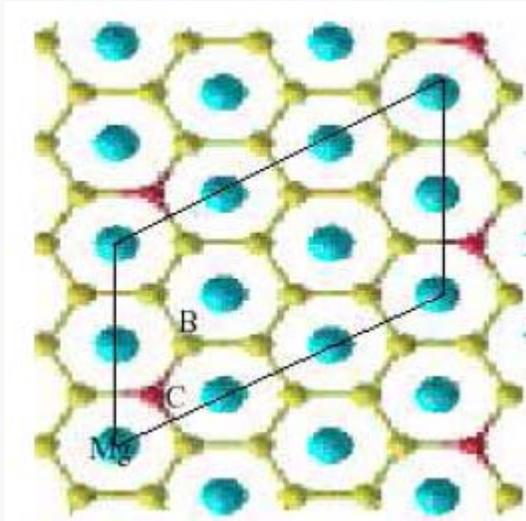
- ◇ **MgB<sub>2</sub>**: a real possibility for a high field conductor, due to high parallel and perpendicular critical fields
- ◇ **Mg<sub>1-y</sub>Al<sub>y</sub>B<sub>2</sub>**:
  1. up to  $y=0.10$ ,  $T_c$  decreases smoothly
  2.  $0.10 < y < 0.25$ , two phase behavior (two  $c$  parameters)
  3.  $y=0.25$ , back to single phase with vanishing  $T_c$
- ◇ **Mg(B<sub>1-x</sub>C<sub>x</sub>)<sub>2</sub>**:
  1.  $dT_c / dx = 1K / \% C$
  2.  $H_{c2}$  increases strongly with C content. ( $\sim 33-35$  T)
  3. It remains the 2-band gap superconductor for  $10 \pm 2\%$  C substitution for B.

# Computational Method

## LAPW (Wien2K)

P. Blaha et al., Comput. Phys. Commun. 59, 399 (1990)

- ◇ **VCA** (10% C doping)
- ◇ **2x3 Supercell method**: ordered impurity calculation



## FPLO

K. Koepnik and H. Eschrig,  
PRB 59, 1743 (1999)

<http://www.ifw-dresden.de/agtheo/FPLO/>

- ◇ **Coherent Potential Approximation (CPA)**: disordered (randomly substituted) alloy calculations

K. Koepnik, B. Velicky, R. Hayn,  
and H. Eschrig,  
PRB 55, 5717 (1997)

# CPA detail

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## ◇ Goal

- 1) determine the filling of the  $\sigma$  band hole states
- 2) obtain the broadening (and potentially splitting) of bands

◇ Doping level:  $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$

$x$	Goal
0.0001	“perfect crystal” reference for evaluation of <b>the numerical algorithms in CPA</b>
0.0833	to compare with $x=1/12$ supercell calculation
0.10 & 0.20	Representative of the system toward the achievable upper range

- ◇  $k$ -mesh: 45x45x10 (1152 irreducible  $k$ -point)
- ◇ Valence orbitals: 2s2p3s3p3d (Mg), 2s2p3d (B&C)

# Smearred band structure (Bloch spectral density)

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- ◇ For ordered single particle spectra,

$$A_k^{bl}(\omega) = \sum_n \delta(\omega - \varepsilon_{kn})$$

$\left( \begin{array}{l} \text{if } \omega = \varepsilon_{kn}, \text{ black dot} \\ \text{otherwise, white} \end{array} \right) \Rightarrow$  usual band structure

- ◇ In disorder case,

$$A_k^{bl}(\omega) = \text{Tr} \left( S(k) \Gamma(k, \omega) \right)$$

where  $S(k)$ : overlap matrix

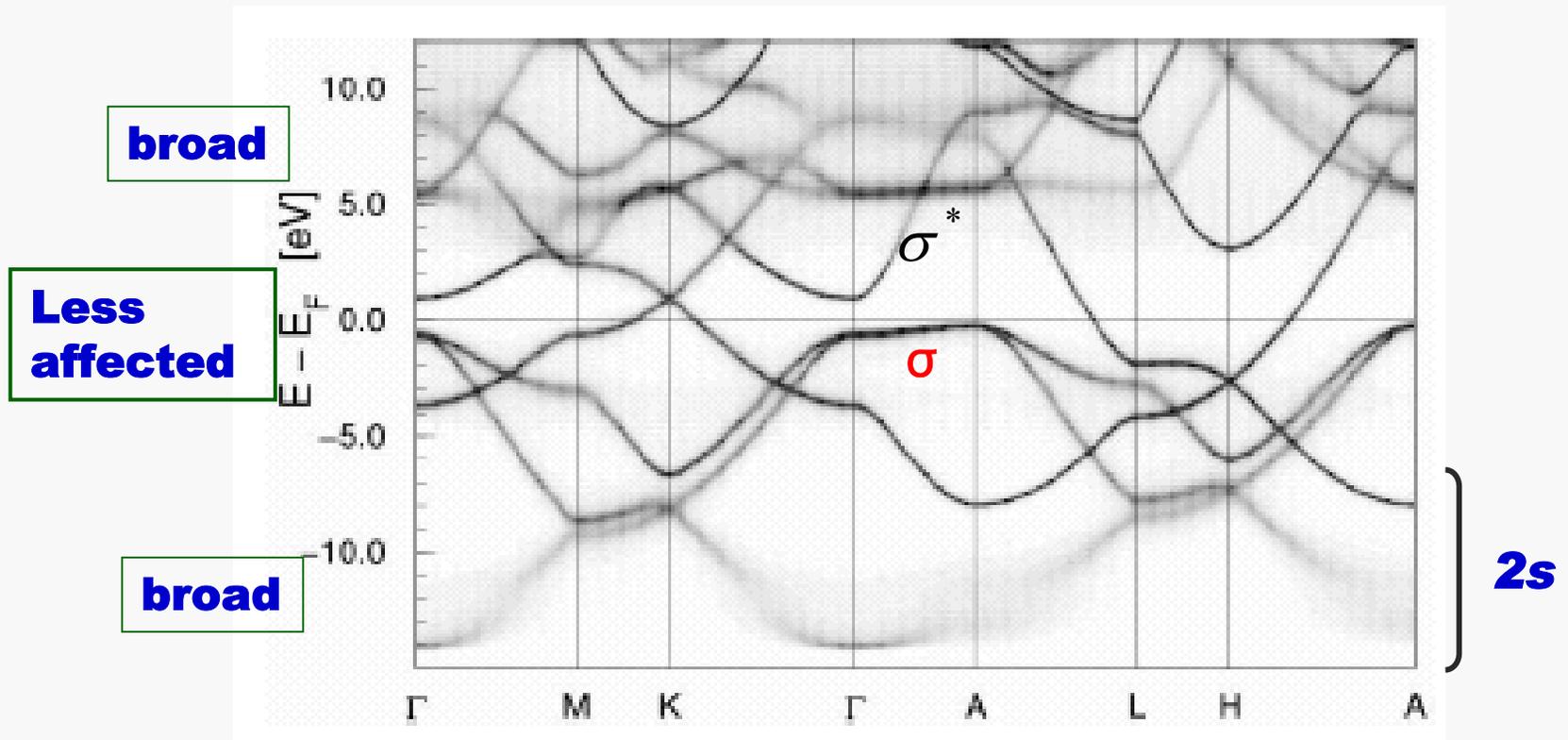
$\Gamma(k, \omega)$ : nonstochastic nonlocal coherent Green function

∴ large A : dense darkness

small A: light

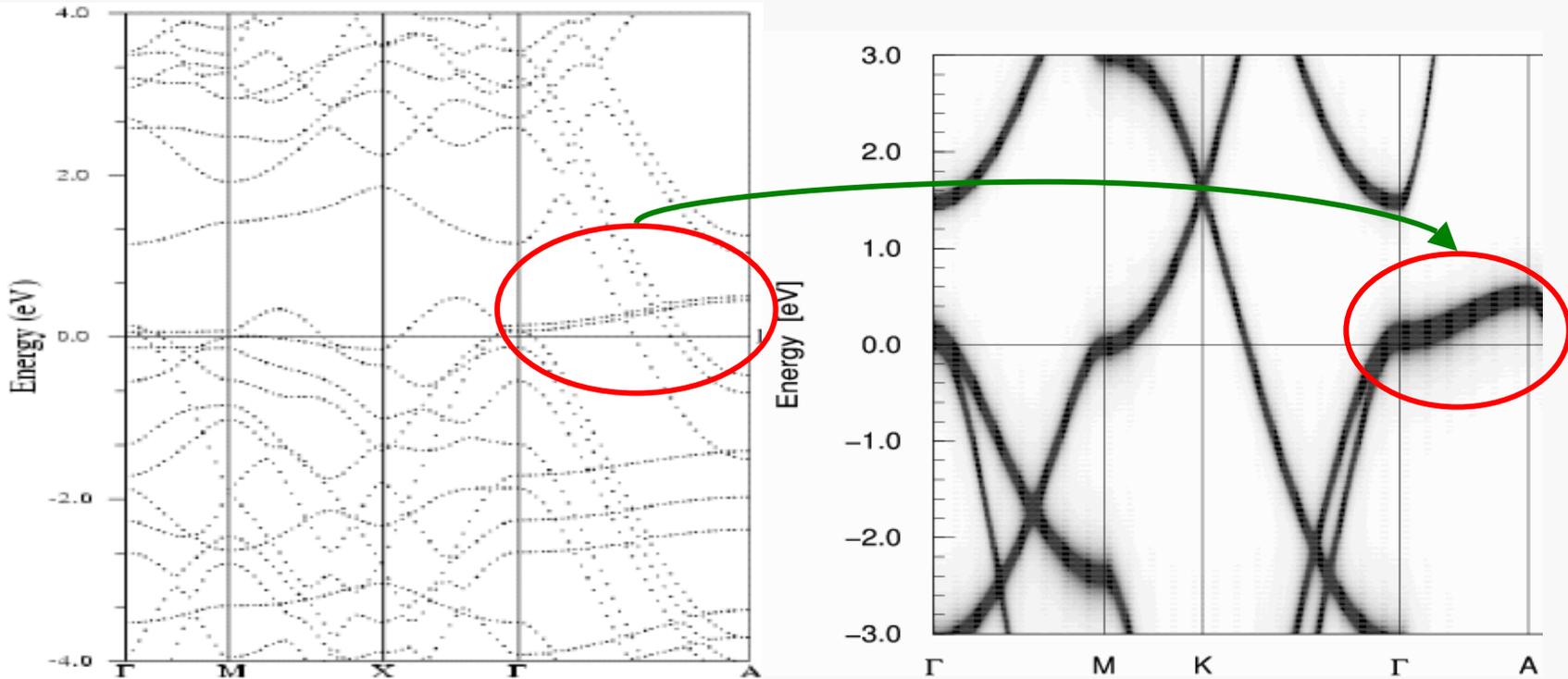
intermediate region: continuous gray gradients

# CPA I ( $x = 0.20$ )



- ◇ Largest broadening ( $2s$ ): C  $2s$  state is noticeably lower in energy leading to increased smearing.
- ◇ Near  $E_F$ , less affected by the chemical disorder.
- ◇ The  $\sigma$  band holes are completely filled.  
⇒ The two-gap superconductivity disappears.

# 2x3 Supercell & CPA ( $x=1/12$ )

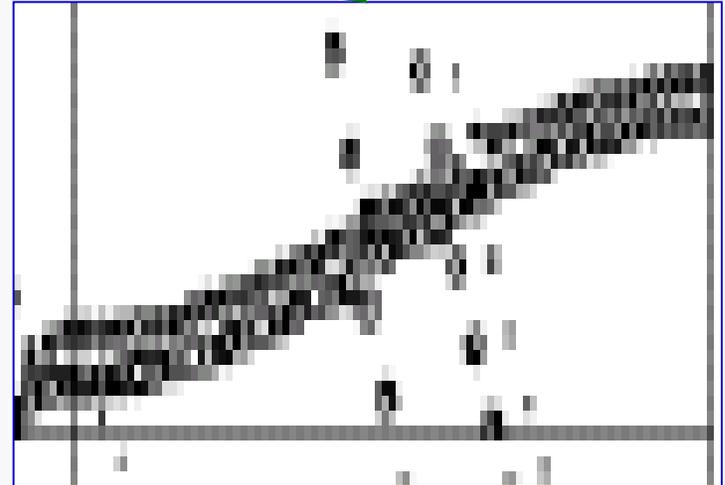
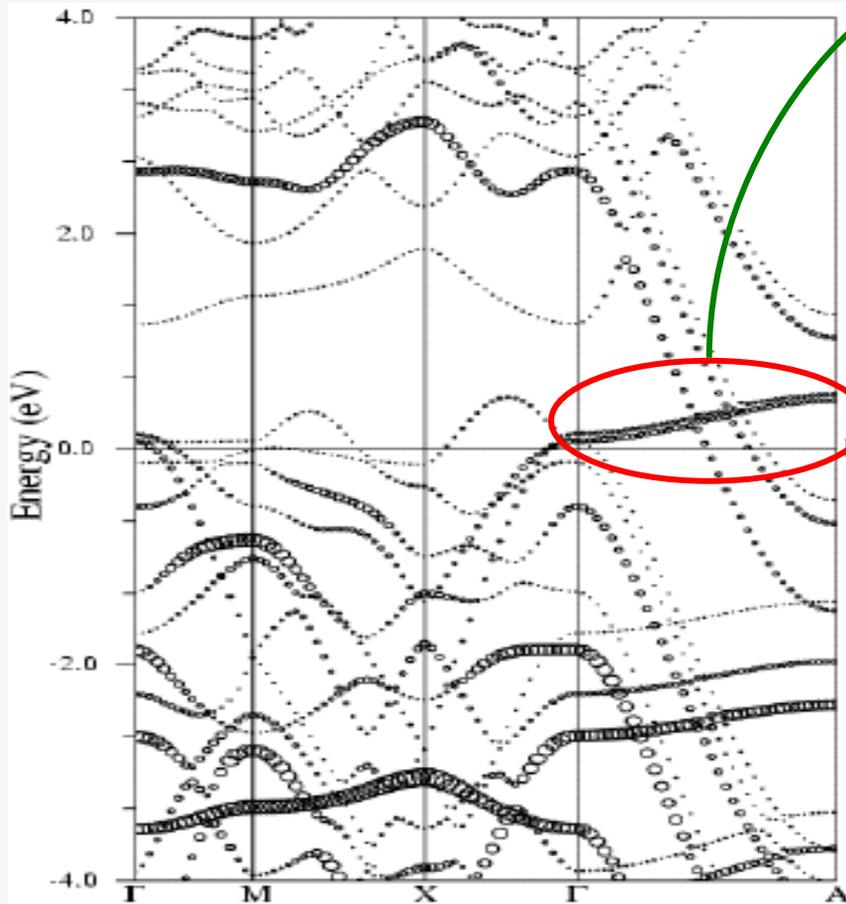


- **C substitution**

1. a change in the average potential in the B-C layer
2. the breaking of symmetry by C replacement of B in the supercell

- **the splitting of the  $\sigma$  bands along  $\Gamma$ -A by 60 meV**

*continued*

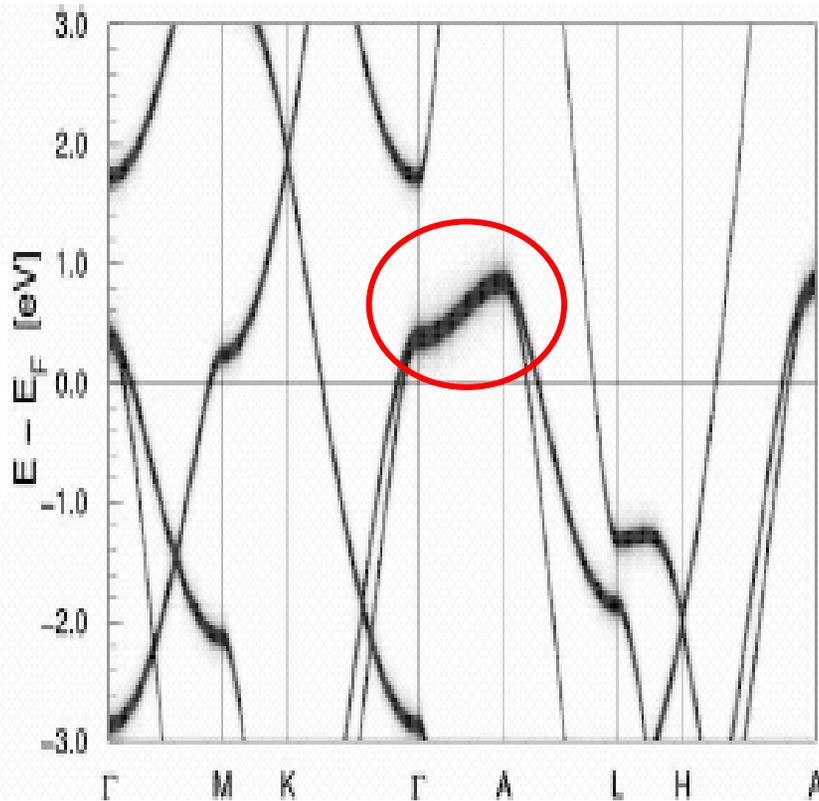


Fatband of the C  $2p$  character

- Lower band is pulled down due to the stronger potential of the C atom than that of the B atom.
- The splitting provides an energy scale for  $\sigma$ -band broadening  $\gamma_0 \approx 7 \text{ meV}/\%C$ .

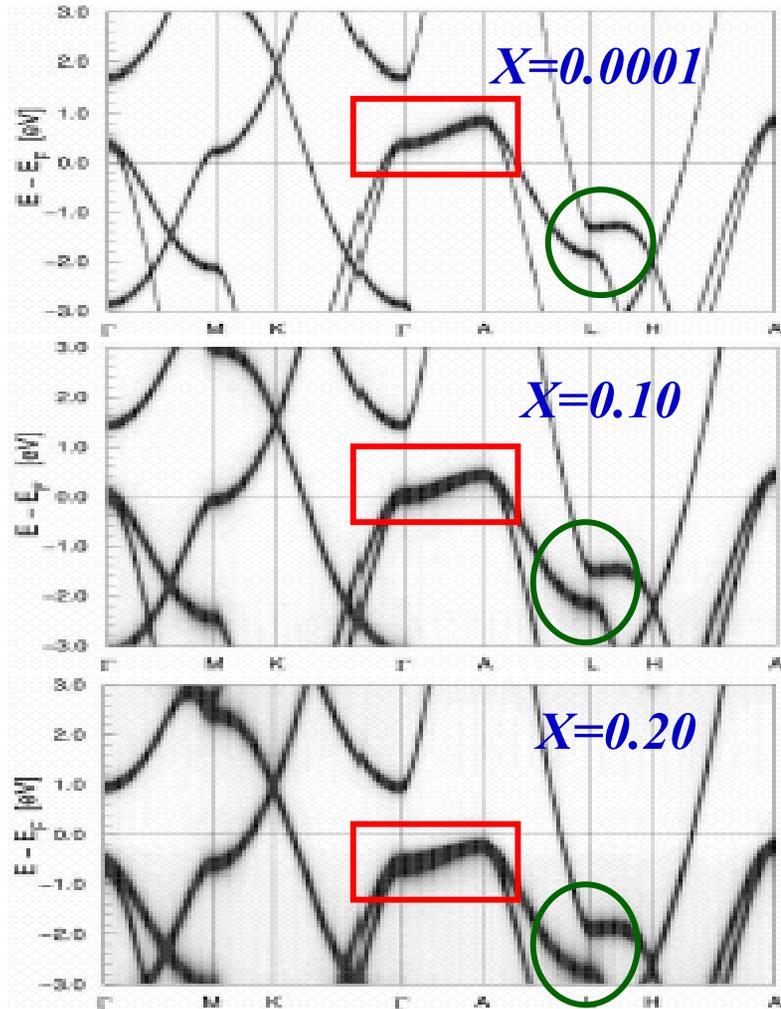
# CPAlI ( $x=0.0001$ )

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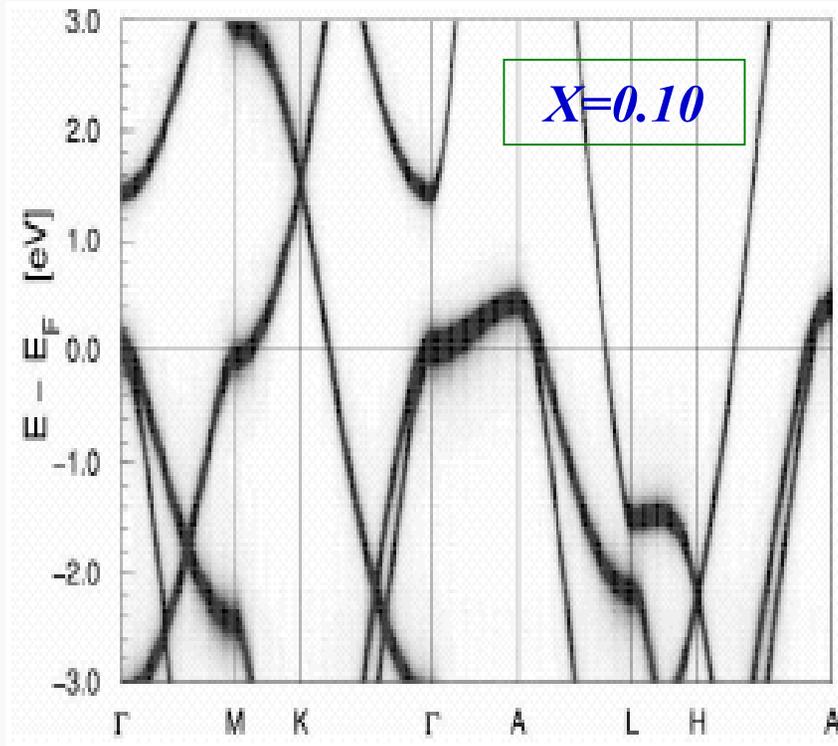
- ◇ Since the numerical algorithms cannot reproduce the  $\delta$ -function bands for  $x \rightarrow 0$ , it is included as a reference for the algorithmic contribution to the width.
- ◇ **Width:**
  - algorithmic contribution
  - + disorder effect
  - + (order effect)

# CPA III (Width of $\sigma$ band)



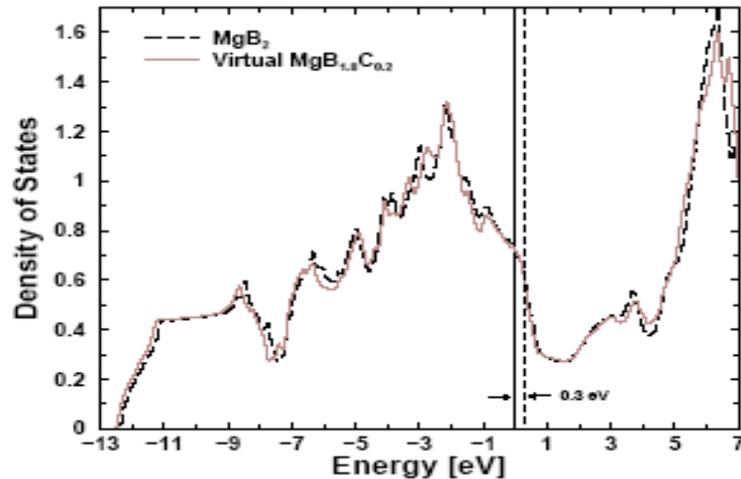
- ◇  $\delta E_F = -0.4$  eV for  $x=0.10$   
-1.0 eV for  $x=0.20$
- ◇ The band shift is not entirely rigid.
  - ← The two valence bands at the L point split apart and broaden with increasing C.
- ◇ averaged  $\gamma \approx 0.21$  eV
  - 0.06 eV from the B/C on-site energy difference
  - 0.15 eV from disorder itself

# CPA IV (mean free path)



- ◇ The width corresponds to a width in wavevector given by  $\gamma = v_F \delta k$ .  
 $\therefore l_F = 2\pi v_F / \gamma$
- ◇ At  $x=0.10$ ,  $l_F$  may vary considerably over the FS due to the anisotropy of the  $v_F$ .
- ◇ At  $\Gamma$ , the cylinder radius shrunk to a point and the very small z-component of  $v_F$ .  
 $\Rightarrow$  a very small  $l_F$  in the z-direction of the order of the layer spacing  $c$ .

# VCA ( $x=0.10$ )

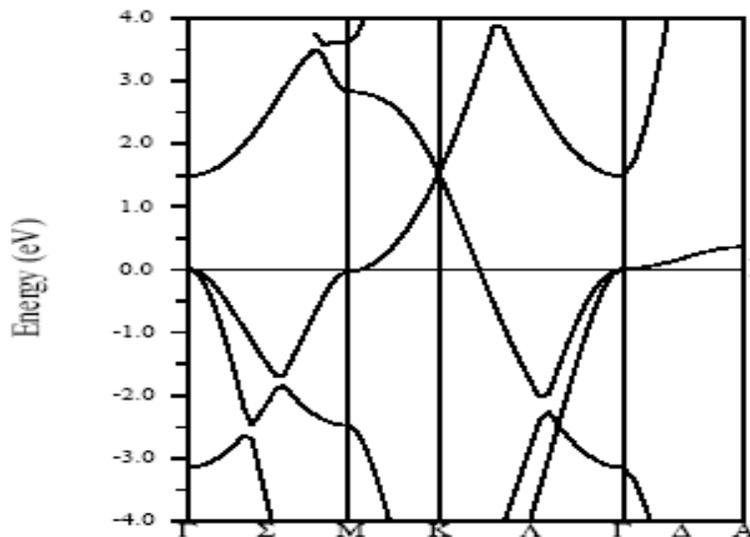


◇ 1% increase in the occupied bandwidth

◇ The raising of E<sub>F</sub> by 0.3 eV

◇ The cylindrical FS radii of the  $\sigma$  bands at  $\Gamma$  vanish.

⇒ corresponding to a topological transition



# $\sigma$ hole concentration

	No. of holes (holes/cell)
$\text{MgB}_2$	0.11
8.33% doping $\text{Mg}_6\text{B}_{11}\text{C}$ supercell	0.070
10% doping $\text{MgB}_{1.8}\text{C}_{0.2}$ Virtual crystal	0.0463
8.33% doping $\text{MgB}_{1.833}\text{C}_{0.167}$ extrapolation	0.057

- ◇ Both the  $\sigma$  band FS are still intact, consistent with the 2-band superconductivity with substantial  $T_c$  as seen in experiments.
- ◇ VCA is not reliable for substitutional C.  
⇒ C cannot be thought simply as “B +  $e^-$ ”.

# Conclusion

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- ◇ The  $\sigma$  band holes begin to disappear rapidly for  $x > 0.10$ .
- ◇ CPA reproduces the  $\sigma$  filling well, while VCA overestimates it.
- ◇ The largest disorder occurs in the  $2s$  region in the lower valence band.
- ◇ As the  $\sigma$  band fills, there will be very strong deviation from “business as usual” in the coupled el.-ph. system.  
(The strength of coupling of bond-stretching modes with  $Q < 2k_F$  continues to increase.)
- ◇  $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$  as well as  $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$  is a good system to study the evolution of the unusual el.-ph. Coupling character.