INTERNATIONAL ATOMIC ENERGY AGENCY
UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
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UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION

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SMR. 628 - 10

Research Workshop in Condensed Matter, Atomic and Molecular Physics
(22 June - 11 September 1992)

Working Party on "DISORDERED ALLOYS"

(24 August - 4 September 1992)

" FERROMAGNETIC ALLOYS "

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AUTUMN TERM - DEPARTMENTAL COLLOQUIUM

5th December 1990

Compositional Orders

Sub-femtoscopic Physics - the High Energy Frontier

(Staunton)
Dr J B Dainton

Department of Physics

University of Liverpool

High energy physics is concerned with the nature of the universe at the most fundamental level. In the last two decades it has taken great strides towards an understanding of the constituent structure and dynamics of matter and energy over the shortest spatial and temporal intervals. Experiments nowadays are capable of resolving any deviations from the so called Standard Model of the universe in the form of unexpected phenomena associated with the fundamental quanta - quarks, gluons, leptons, vector bosons - and their interactions, quantum chromodynamic and electroweak. The status of these experiments and their tests and searches for new phenomena will be outlined and discussed in a non-specialist way.

Seminar to take place at 4.30pm in the
Physics Department Colloquium Room - P523
for further information contact Dr C F McConville (R428)

1.

Compositional Order in Metallic Alloys

JBS

D. D. Johnson   Sandia Nat'l Labs, U.S.A

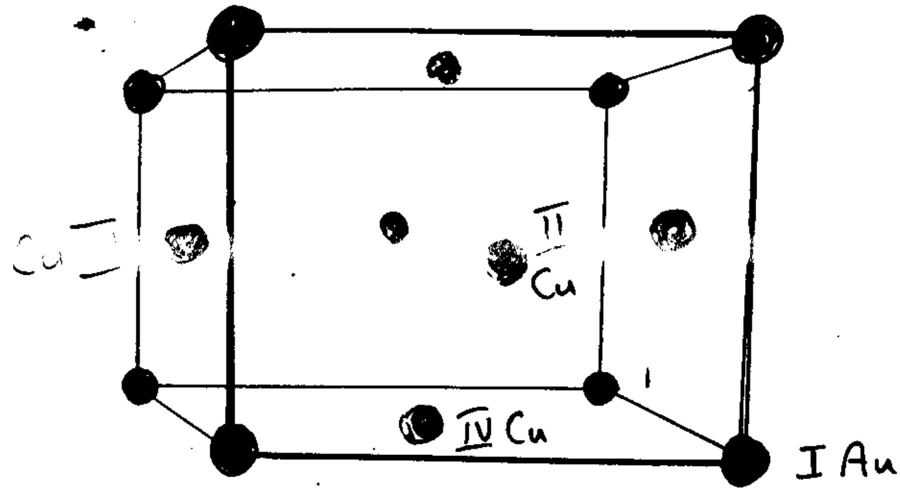
F. J. Pinski   Univ. of Cincinnati "

Electronic "glue" is key to understanding properties of metallic alloys

Compositional order, phase stability

Theorists   Experimentalists   Metallurgists

2.



Many 2 component (A, B) random solid solution. Lattice randomly occupied

$T\downarrow$ Phase separation or ordering

$Cu_c\, Au_{1-c}$ $\quad c \sim .75$

High temperature f.c.c., $c_I = c_{II} = c_{III} = c_{IV}$

$T < T_c$ $\quad c_I < c_{II} = c_{III} = c_{IV}$

Order parameter $\eta = c_I - c_{II}$

3. Introduce occupation variable (label)

$$\xi_i = \begin{cases} 1 & \text{if A atom is on } \underline{R}_i \\ 0 & \text{if B atom is on } \underline{R}_i \end{cases}$$

$$c_i = \langle \xi_i \rangle$$

In random solid solution $c_i = c$

Symmetry broken $\rightarrow \{c_i\}$ different from others. Concentration wave description

$$c_i = c + \frac{1}{2} \sum_{\nu} \left(c_\nu e^{i \underline{q}_\nu \cdot \underline{R}_i} + c_\nu^* e^{-i \underline{q}_\nu \cdot \underline{R}_i} \right)$$

Ordered structures described by a few $\underline{q}_\nu$.

e.g. Cu_3Au ($L1_2$)

$$\underline{q}_\nu = 2\pi/a\,(1,0,0),\ \frac{2\pi}{a}(0,1,0) \text{ and } \frac{2\pi}{a}(0,0,1)$$

$$c_{I} = c - \tfrac{1}{4}\eta, \quad c_{\underline{II}} = c_{\underline{III}} = c_{\underline{IV}} = c + \tfrac{3}{4}\eta$$

Tendency to order → occupational correlation function

$$\alpha(q) = \beta \sum_j \left(\langle \xi_i \xi_j \rangle - \langle \xi_i \rangle \langle \xi_j \rangle \right) e^{i q \cdot (R_i - R_j)}$$

Peaks at values of q_ν describing likely ordered structure.

Landau Ginzburg theory
(Landau, Lifshitz, Khachaturyan ...)

Lifshitz "special points" — symmetry grounds

f.c.c. $(0, 0, 1), \ldots$
$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}), \ldots$
$(\frac{1}{2}, 1, 0) - \ldots$

b.c.c. $(0, 0, 1)$
$(\frac{1}{2}, \frac{1}{2}, 0)$
$(\frac{1}{2}, \frac{1}{2}, 0)$

"Special points" generate most commonly observed ordered phases

Non special point structures . . .
(incommensurate, layered structures etc)

Lifshitz special points:

Ordering when $\mathcal{S}(\vec{k})$ largest

$\therefore$ Find $\nabla_{\underline{k}} \mathcal{S} = 0$ (max.)

1.) system dependent details of $\mathcal{S}(\underline{k})$

2.) from symmetry of disordered phase alone $\leftarrow$ Lifshitz special points

Group $g(\underline{k})$ containing operations which bring lattice in coincidence with itself and $\underline{k}$ unchanged

— subgroup of space group of disordered phase.

$\nabla_{\underline{k}} \mathcal{S}$ also invariant

But if 2 or more symmetry operations of point group $g(\underline{k})$ intersect at one point, then orientation of $\nabla_{\underline{k}} \mathcal{S}$ alters

Resolution $\longrightarrow$ $\nabla_{\underline{k}} \mathcal{S} \equiv 0$

- Why do some systems form ordered structures but others phase segregate? $A_c B_{1-c}$

- What determines type of ordered structure?
  ...... metallurgical properties etc.
  ...... Alloy design

Hume-Rothery rules: (1930's)

(i) Electron per atom ratio
band filling **Magnetic state.**

(ii) Size effect
15% rule
"Big" & "small" atoms order

(iii) Electronegativity
.... charge transfer

↳ Detailed understanding in terms of behaviour of electron glue

Bragg - Williams model:

$$H = -\tfrac{1}{2}\sum_{ij}\Big(V_{ij}^{AA}\xi_i\xi_j + V_{ij}^{AB}\xi_i(1-\xi_j) + V_{ij}^{BA}(1-\xi_i)\xi_j + V_{ij}^{BB}(1-\xi_i)(1-\xi_j)\Big)$$

$$= -\tfrac{1}{2}\sum_{ij} w_{ij}\,\xi_i\xi_j - \tfrac{1}{2}\sum_i w_i^{(1)}\xi_i - w^{(0)}$$

$$w_{ij} = V_{ij}^{AA} + V_{ij}^{BB} - 2V_{ij}^{AB}$$ interchange energy

$$Z = \sum_{\{\xi_k\}} e^{-\beta\left(H\{\xi_k\} - \nu\sum_i(\xi_i - \frac{1}{2})\right)}$$

$$F = -\tfrac{1}{\beta}\ln Z$$

$$c_i = \frac{1}{Z}\sum_{\{\xi_k\}}\xi_i\, e^{-\beta\left(H - \nu\sum_k(\xi_k - \frac{1}{2})\right)}$$

Feynman-Peierl's Inequality

$$F < F_0 + \langle H - H_0 \rangle_0$$

$\Rightarrow$ Mean field theory

$$H_0 = \sum_i h_i^{eff} \xi_i$$

$$h_i^{eff} = -\sum_j w_{ij} c_j - w_i$$

$$c_i = \frac{e^{-\beta(h_i^{eff} - \nu)}}{(e^{-\beta(h_i^{eff} - \nu)} + 1)}$$

High T, $c_i = c$.

Correlations, apply $\{\nu_i^{ext.}\}$
calculate $\{\delta c_i\}$

$$\delta c_i = \beta c(1-c) \sum_j (w_{ij} \delta c_j + \nu_i^{ext})$$

$$\rightarrow \alpha(\underline{k}) = \sum_j \frac{\delta c_i}{\delta \nu_j^{ext}} e^{i\underline{k}\cdot(\underline{R}_i - \underline{R}_j)} = \frac{\beta c(1-c)}{(1 - \beta c(1-c) w(\underline{k}))}$$

2.

5. Theory of Compositional Ordering in Alloys.

— Concentration Waves (Khachaturyan
— Electronic basis (Gyorffy & Stocks
(Ducastelle et al.
(de Fontaine ....

Ω for configuration of nuclei (S.D.F.)

— $\Omega\{\xi_i\}$

$\xi_i = 1$ i^{th} site occupied by A atom

$= 0$ " " " B "

$c_i = \langle \xi_i \rangle$

c_i can be different on every site.

$c_i = c$ $\longrightarrow$ SCF-KKR-CPA

$\bar{\Omega}$, ρ_A, ρ_B, ... t_c, $\tau^{c,00}$

SDF. $\rightarrow$ $\dfrac{\delta \bar{\Omega}}{\delta \rho_A(\xi_i)} = \dfrac{\delta \bar{\Omega}}{\delta \rho_B(\xi_i)} = 0$

3.
6. Averaging over configurations

$$F = -\frac{1}{\beta} \ln \prod_i \sum_{\xi_i = 0,1} \exp\Big(-\beta \Omega\{\xi_i\} - \nu \sum_i (\xi_i - \tfrac{1}{2})\Big)$$

Feynman - Peierls Inequality

$$F \leqslant F_0 + \langle \Omega - \Omega_0 \rangle^{0} = \tilde{F}$$

Choose $\Omega_0 = \sum_i f_i(\xi_i)$ .... $\langle \ldots \rangle^{0} = \dfrac{\prod_i \sum_{\xi_i} \cdots e^{-\beta \Omega_0}}{\prod_i \sum_{\xi_i} e^{-\beta\Omega_0}}$

$$c_i = \frac{\exp\left(-\beta[f_i(1) - f_i(0)] + \beta\nu\right)}{\left[\exp\left(-\beta(f_i(1) - f_i(0)) + \beta\nu\right) + 1\right]}$$

$$f_i(\xi_i') = \langle \Omega \rangle^{0}_{\xi_i = \xi_i'}$$

Inhomogeneous C.P.A.

$$c_i \tau^{A,ii} + (1 - c_i)\tau^{B,ii} = \tau^{c,ii}$$

$$\tau^{c,ii} = t_{c,i} + \sum_{k \neq i} t_{c,i} G^{0}_{ik} \tau^{c,ki}$$

4. Ordering i.e. correlations

7.

Apply inhomogeneous external field, calculate response

1. Apply $\sum_i u_i^{ext.}$ which couples to occupation variable ξ_i

$\rightarrow$ induces $\{\delta c_i\}$ but also $\{\delta \rho_i^{A(B)}\}$

$$\rightarrow \quad \alpha_{ij} = \frac{\partial c_i}{\partial(v_j^{ext} - \eta)} = \beta\left(\langle \xi_i \xi_j \rangle - \langle \xi_i \rangle \langle \xi_j \rangle\right)$$

Lattice F.T.

$$\alpha(q) = \frac{\beta\, c(1-c)}{\left(1 - \beta\, c(1-c)\, \mathcal{S}(q)\right)}$$

$\mathcal{S}(q)$ Krivoglaz-Moss-Clapp

$$\mathcal{S}_{ij} = V_{ij}^{AA} + V_{ij}^{BB} - 2V_{ij}^{AB}$$

Interchange

(3. Apply $\sum V_i^{ext}$ which couples to ρ_i

$\rightarrow$ charge density correlations)

B.
9
14

$$\alpha(q) = \frac{\beta c(1-c)}{\left\{1 - \beta c(1-c)\left[S^{(2)}(q) + \delta_1(q) + \delta_2(q)\right]\right\}}$$

band filling + splitting
Fermi Surface,
only term if
charge effects
neglected.

"local" charge
relaxation
effects.
Structure in
q-space follows
$S^{(2)}(q)$

"Madelung"
Charge Transfer
piece

$$\sim \frac{(\Delta Q)^2 \tilde{G}(q)}{(1 + \chi(q)\tilde{G}(q))}$$

screening

ΔQ = charge transfer
$= (\rho_A - \rho_B - Z_A + Z_B)$

Electronic structure $\longleftrightarrow$ "Madelung"

8.

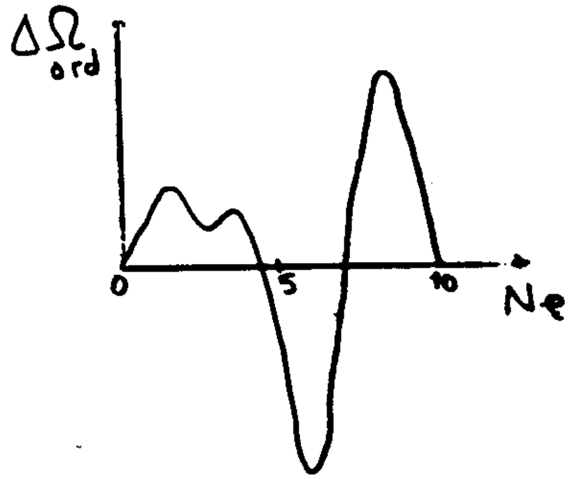
Factors that effect tendency to order (ingredients of $S(q)$)

— Hume-Rothery

electrons/atom,... band filling, electronegativity atomic sizes ("big" & "small" atoms tend to order.)

T-B analyses band filling

$$\Delta\Omega_{ord.} = \Omega_{order} - \bar{\Omega}$$



$c = .25$

$\frac{\varepsilon_A - \varepsilon_B}{W} \sim .5$

↳ Half-filled bands --- ordering
Nearly filled/empty band -- clustering

11. -
18.

Ni$_{50}$Pt$_{50}$

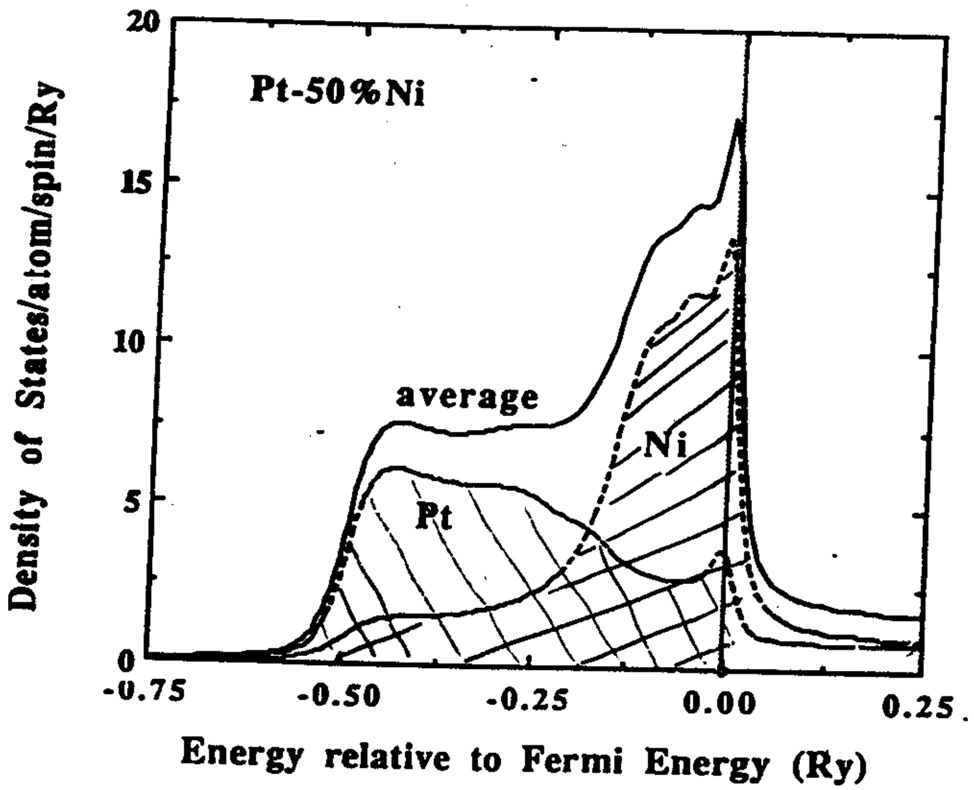


Figure 2.

Cu - Pd Fermi Surface

$Ni_{50}Pt_{50}$ $\Delta Q \sim 0$

$$\alpha(\vec{q}) = \frac{\beta c(1-c)}{(1-\beta c(1-c)\, S^{(2)}(\vec{q}))}$$

off-diagonal disorder
Pinski et al
PRL (1991)

$(q_z = 0.0)$

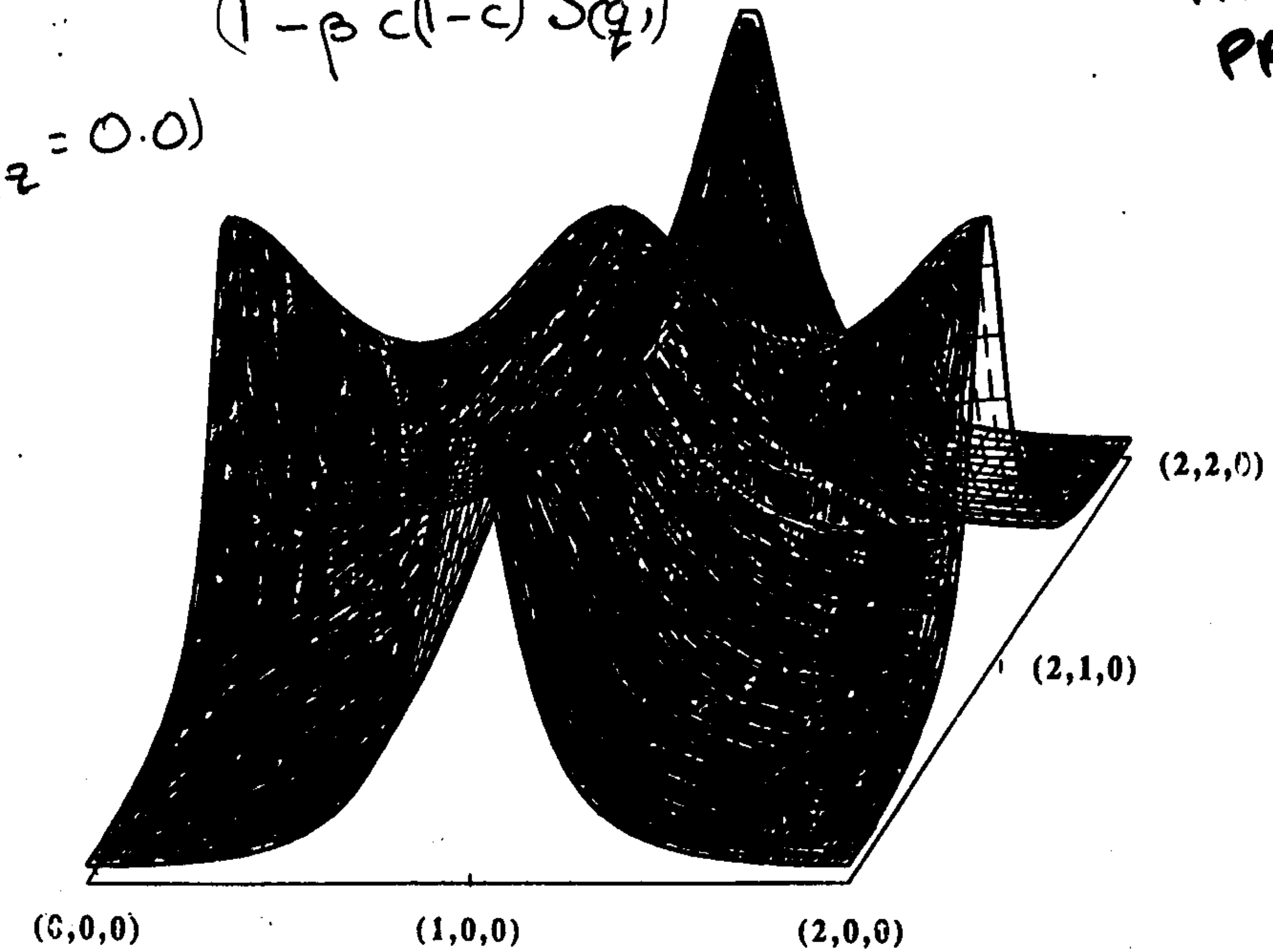


Figure 1.

Neglecting charge effects, studied

$Ni_{75}V_{25}$, $Pd_{75}V_{25}$, $Co_{75}Ti_{25}$

$\downarrow$ $\downarrow$ $\downarrow$

$L1_2$ $L1_2$ $L1_2$

$(q = 0,0,1)$

DO_{22} DO_{22} $L1_2$ Expt.

$(q = 0, 1/2, 1)$

Nice experimental data for

$Cr_{11}Ni_{89}$, $Cr_{20}Ni_{80}$

$\rightarrow$ DO_{22} correlations $(q = 0, 1/2, 1)$

subsidiary peak at $(2/3, 2/3, 0)$

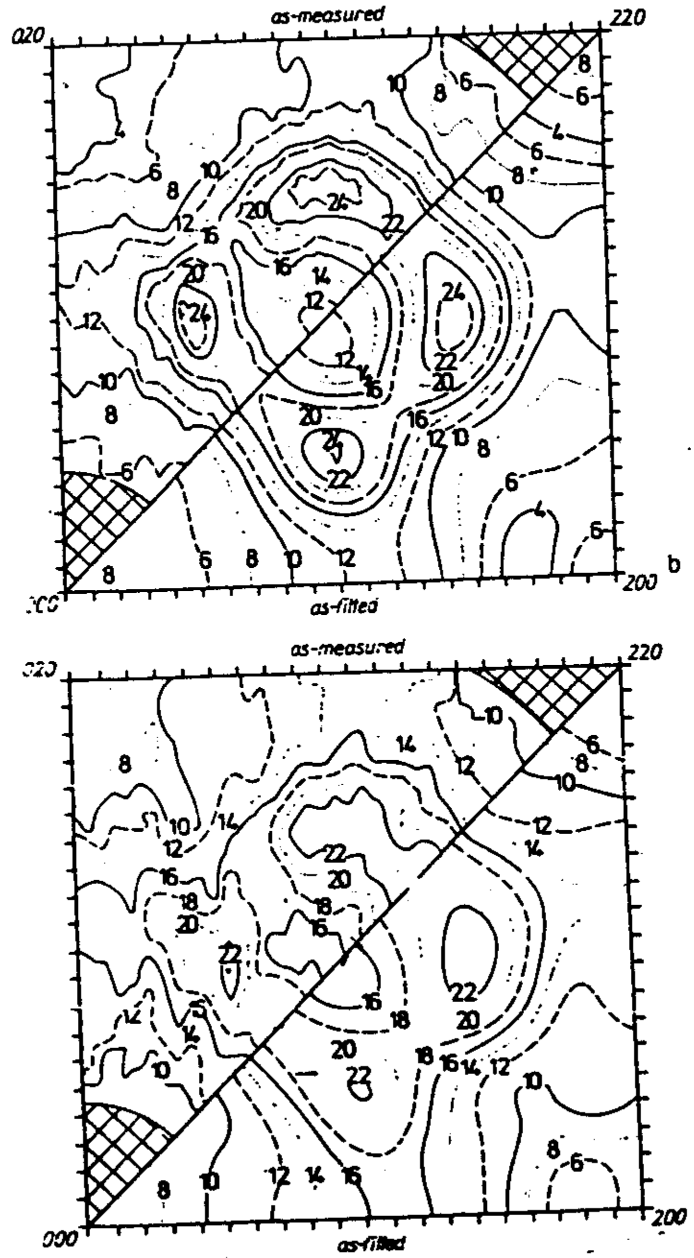
$CrNi_2$

ibution, a coarser

m DATAP of the
ption coefficients
le scattering was
in overall Debye-
0.291×10^{-2} nm²
um $B = 0.485 \times$
= 10.3(1) fm and
ions were σ^{r}_{inc} =
$^{Ni}_{inc}$.

cal (100) plane is
ation with ageing
e maxima at posi-
, the maxima of
|, start to merge.
dicating that dis-

weak wings of the
ound Bragg reflec-
t beam) were not
o the direct beam,



$q_z = 0.0$

Diffuse neutron scattering

$Cr_{20}Ni_{80}$

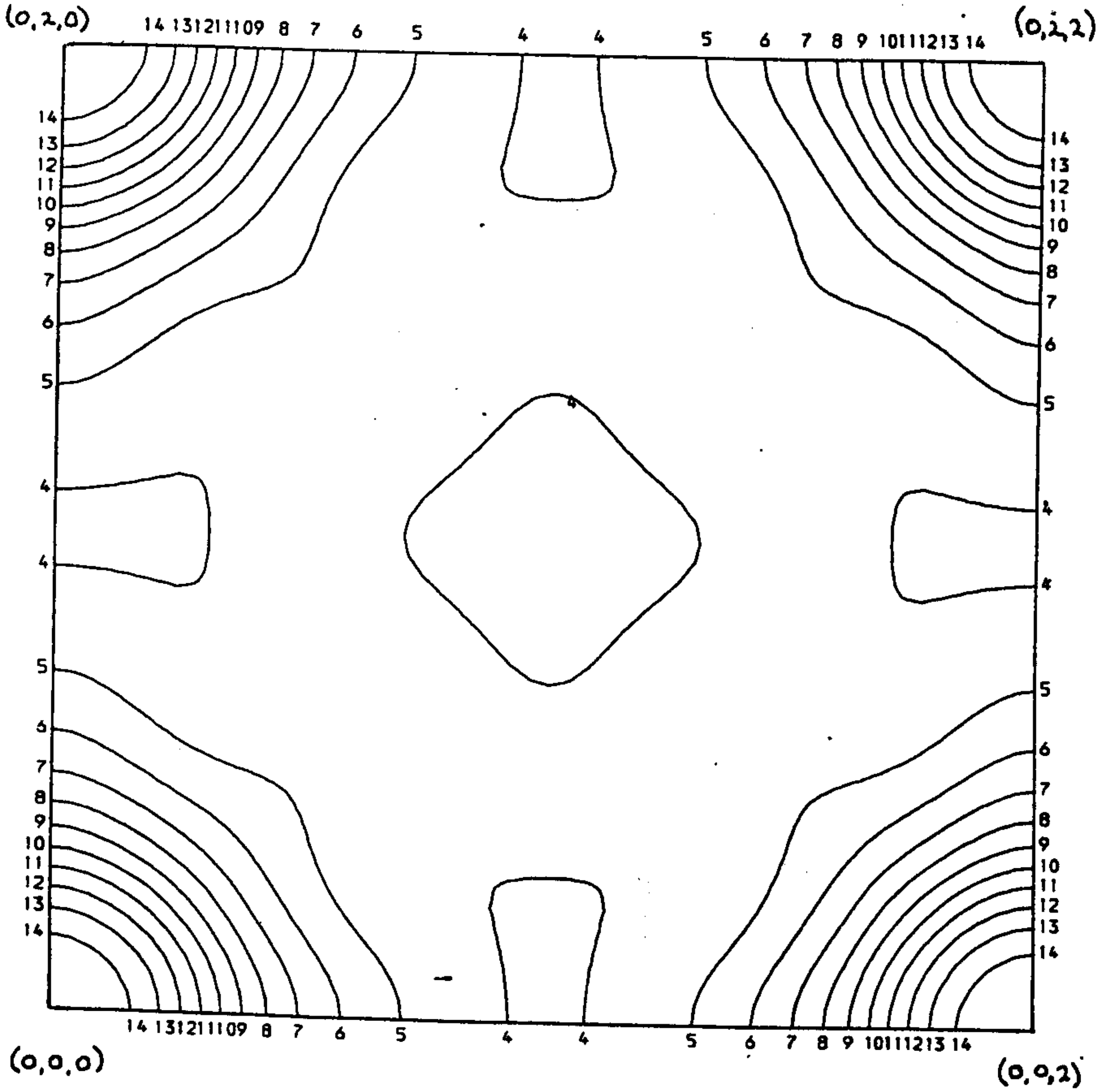
Fig. 1b, c

Fig. 1. Diffuse scattering intensity $I_d(h)$ in 0.1 Laue units (equidistant isointensity lines) for a (100) plane and recalculated using 21 α_{lmn} and 26 γ^i_{lmn} (Tables 1, 2) for a) sample 1, b) sample 2, and c) sample 3

B. Schonfeld, L. Reinhard, G. Kostorz and W. Bührer, Phys. Stat. Sol. (b), 148, 457 (1988) Figure 3.

Neglecting charge transfer

$Cr_{20}Ni_{80}$ .... clustering.



Compositional correlation function, $\alpha(q)$, $q_x = 0.0$ plane for $Ni_{80}Cr_{20}$ without 'charge transfer' effects.

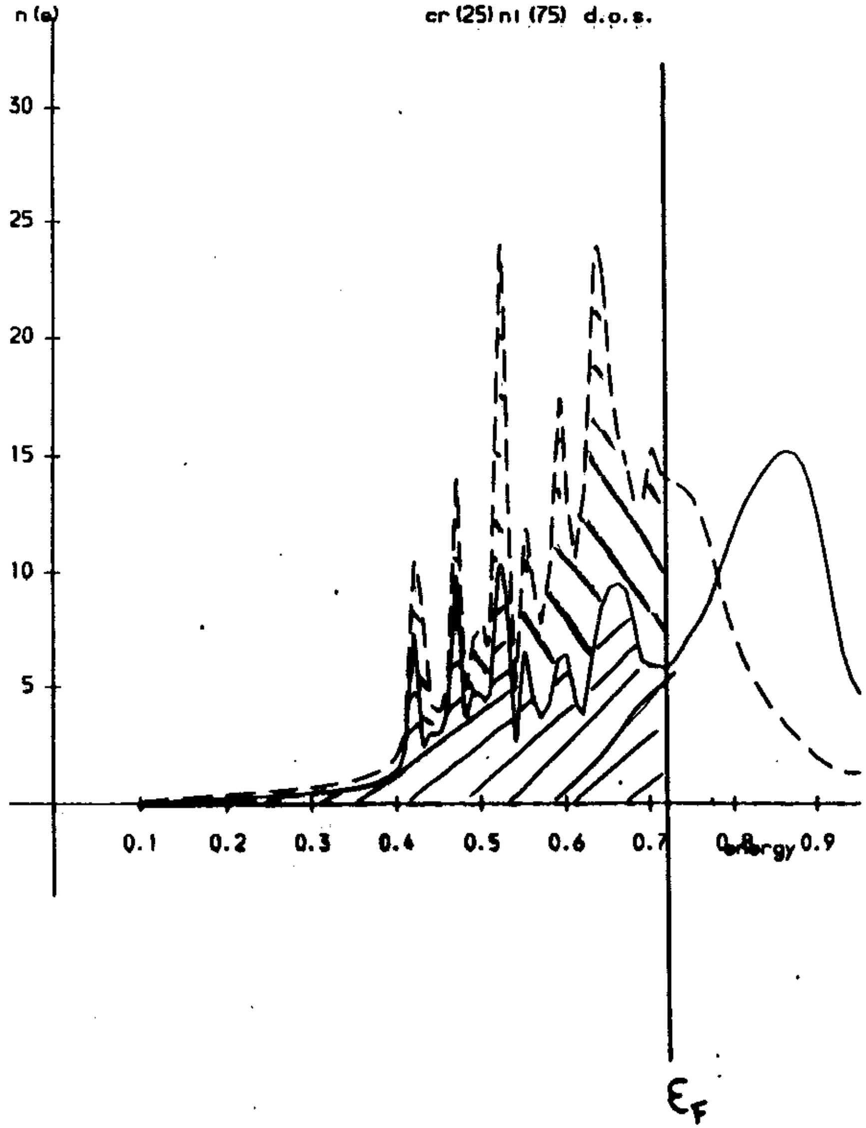
Figure 2.

$$\alpha(q) = \frac{\beta c(1-c)}{(1-\beta c(1-c) S^{(2)}(q))}$$

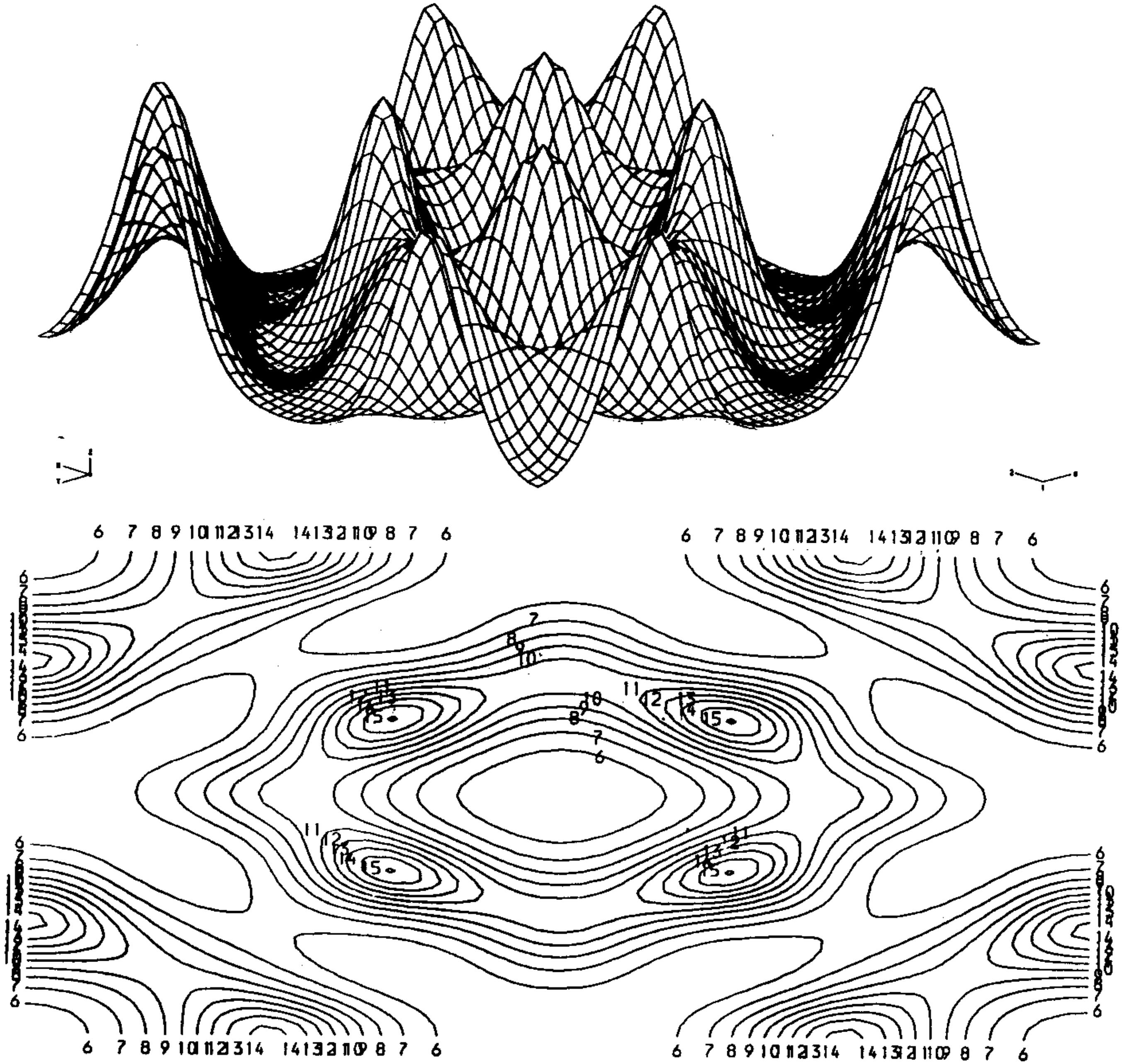
Whoops!

takes over

15.
23.



"Split bands" partially filled — tendency for phase segregation



$Cr_{20}\,Ni_{80}$

$T = 900K$

$q_x = 0.0$

+ Cavity field

$T_c \sim 700K$

Including charge transfer effects

$\Delta Q \sim -.2$ electrons

25.

Hume-Rothery empirical rules for compositional order .... on the way to a 'first principles' basis

(i) electrons/atom, bandfilling (exchange splitting of bands)

(ii) Size effect

(iii) Electronegativity

First calculations involving all – Chromium-Nickel.

<u>Future Work</u>

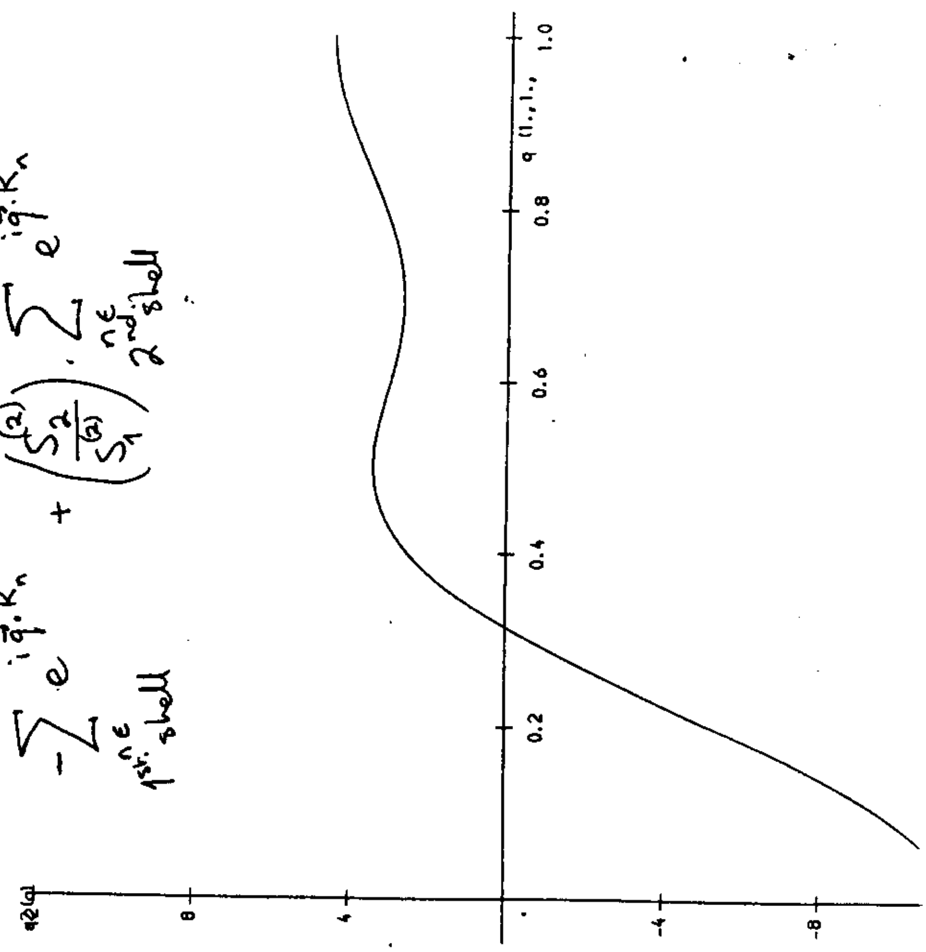
(i) Magnetic Alloys
Interrelation between compositional and magnetic ordering

(ii) Strain Effects
Ti-Al ...

THEORY OF COMPOSITIONAL AND MAGNETIC CORRELATIONS IN ALLOYS

- part I : Effect of magnetic state on compositional ordering

part II : Thermally induced magnetic fluctuations



$-\sum_{1^{st}\,shell}^{n\epsilon} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_2^{(2)}}{S_1^{(1)}}\right)\sum_{2^{nd}\,shell}^{n\epsilon} e^{i\vec{q}\cdot\vec{R}_n}$
0.2
0.4
0.6
0.8
1.0
8
4
-4
-8

1.

- aim of alloy theory — extraction of electronic mechanisms responsible for ordering.

- ∴ theory of correlation functions in terms of alloys' electronic structures.

- Comparison with X-ray + neutron scattering experiments

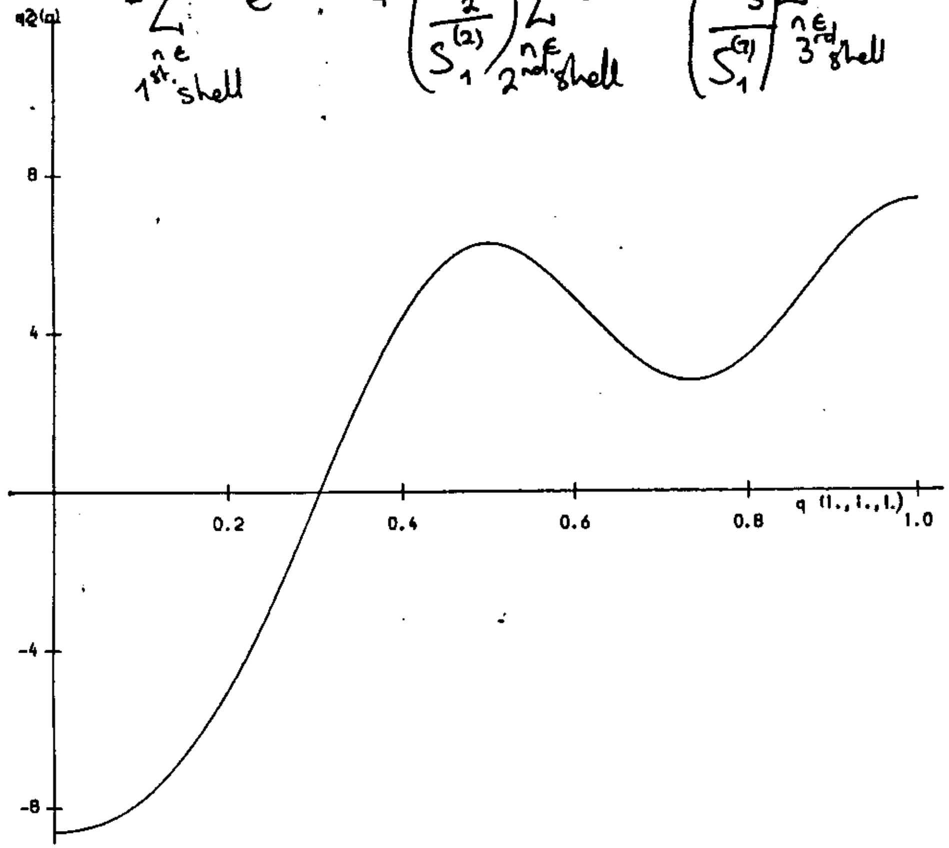
- Here magnetic alloys .....
  ∴ need compositional
  magnetic
  + magneto-compositional
  correlation functions

- Diffuse polarised neutron scattering experiments on single crystals to test theory

..... examples ..... Fe - V,

Ni - Fe alloys

$$-\sum_{\substack{n\in \\ 1^{st}\text{ shell}}} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_2^{(2)}}{S_1^{(2)}}\right)\sum_{\substack{n\in \\ 2^{nd}\text{ shell}}} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_3^{(2)}}{S_1^{(1)}}\right)\sum_{\substack{n\in \\ 3^{rd}\text{ shell}}} e^{i\vec{q}\cdot\vec{R}_n}$$



Starting point

⇒ accurate, 'first principles' treatment of electronic structure of compositionally disordered ferromagnetic alloy $A_c B_{1-c}$

SCF - KKR - CPA.

μ_A, μ_B, bulk modulus, lattice constants etc.

[Multiple scattering framework
→ $t_{A(B)}$, t_c, τ^c]

Slater Pauling Curve

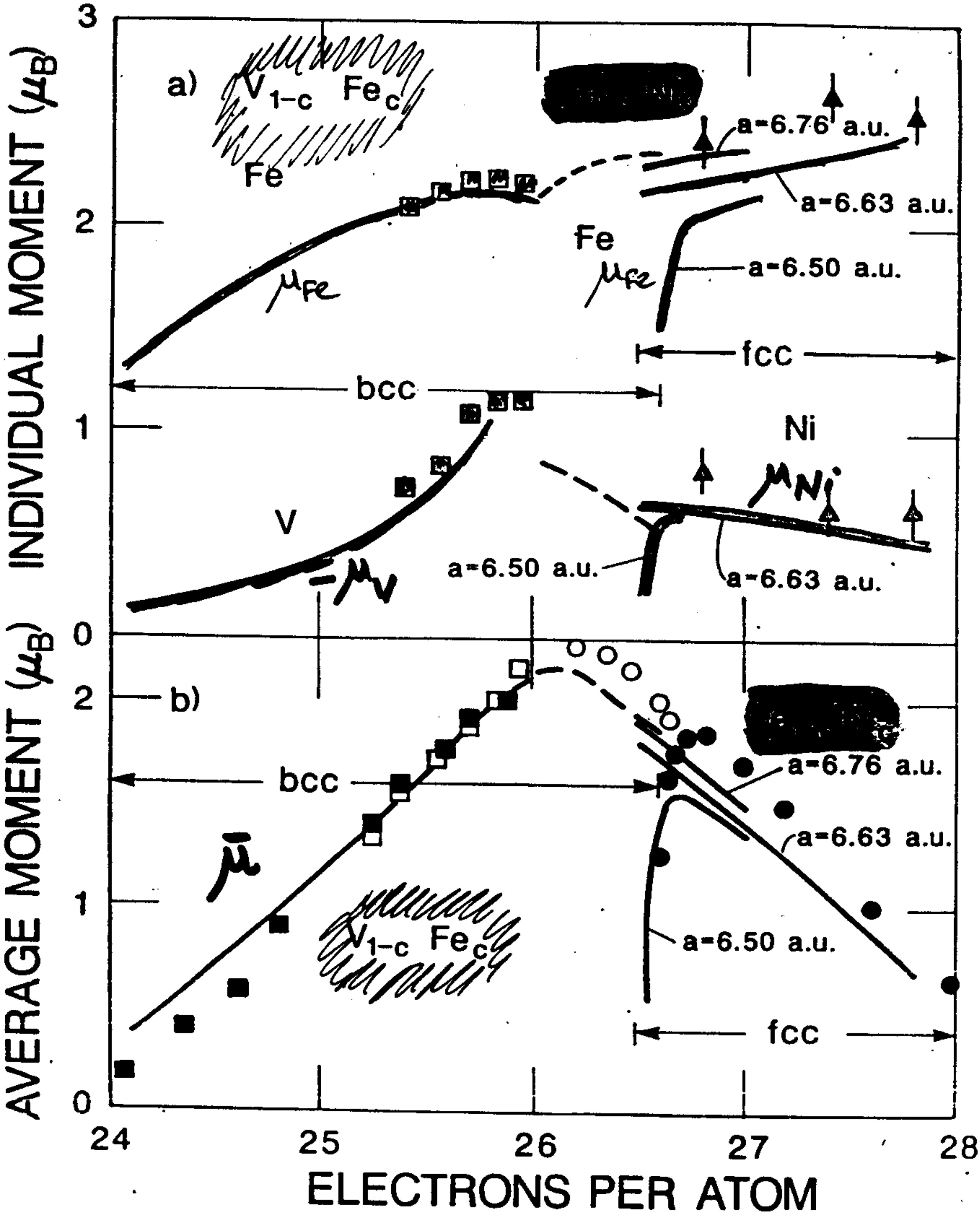


Fig 1

Compositional Order & Magnetic Structure of Transition Metal Alloys

Consider Ω for configuration of nuclei (SDF) — $\Omega\{\xi_i\}$

$\xi_i = 1$ --- i^{th} site occupied by A atom
$= 0$ --- " " " " B "

Allow c_i to vary from site to site $\boxed{c_i = \langle \xi_i \rangle}$ ($c_i = c$ SCF-KKR-CPA)

Averaging over configurations

$$F = -\frac{1}{\beta} \ln \prod_i \sum_{\xi_i = 0,1} \exp(-\beta\,\Omega\{\xi_i\})$$

Feynman – Peierls Inequality

$$F \leq F_0 + \langle \Omega - \Omega_0 \rangle^0 = \tilde{F}$$

Choose $\Omega_0 = \sum_i f_i(\xi_i)$ $\qquad \langle \ldots \rangle^0 = \dfrac{\int \ldots e^{-\beta\Omega_0}}{\int e^{-\beta\Omega_0}}$

$$c_i = \frac{\exp(-\beta f_i(1))}{\left(\exp(-\beta f_i(1)) + \exp -\beta f_i(0)\right)} \qquad f_i(\xi_i') = \langle \Omega \rangle^0_{\xi_i = \xi_i'}$$

Inhomogeneous C.P.A

$$c_i\, \tau^{A,ii} + (1 - c_i)\, \tau^{B,ii} = \tau^{c,ii}$$

$$\tau^{c,ii} = t_{c,i} + \sum_{k \neq i} t_{c,i}\, G^0_{ik}\, \tau^{c,ki}$$

5. Ordering, vs. correlations.

Apply inhomogeneous external fields, calculate response.

1. (i) Apply $\sum_i v_i^{ext}$ which couples to occupation variable ξ_i

Induces $\Rightarrow \{\delta c_i\}$ but also in ferromagnetic alloy $\{\delta \mu_i\}$

(ii) Solve inhomogeneous C.P.A by expanding about homogeneous KKR-CPA case $t_{c,i}^{-1} = t_c^{-1} + \delta t_{c,i}^{-1}$,

$$\tau^{c,ii} = \tau^{c,00} - \sum_j \tau^{c,ij}\, \delta t_{c,j}^{-1}\, \tau^{c,ji}$$

$$\delta t_{c,j}^{-1} = \delta t_{c,j}^{-1}(\{\delta c_i\},\{\delta \mu_i\})$$

Eventually find

$$\alpha_{ij} = \frac{\partial c_i}{\partial v_j^{ext}} = \beta\left(\langle \xi_i \xi_j\rangle - \langle \xi_i\rangle\langle \xi_j\rangle\right)$$

pair correlation function

Lattice Fourier Transform

$$\alpha(\vec{q}) = \frac{\beta\, c(1-c)}{(1-\beta c(1-c)\, S(\vec{q}))}$$

6

$S(\vec{q})$ — from electronic structure (sometimes Fermi Surface) of disordered alloy.

(Gyorffy & Stocks — paramagnetic alloys with long period structures have $S^{(2)}(\vec{q})$ which is dependent on flat pieces of Fermi Surface)

Krivoglaz - Moss - Clapp.

$$S_{ij} = V_{ij}^{AA} + V_{ij}^{BB} - 2V_{ij}^{AB} \quad \text{Interchange}$$
$$+ \left(J_{ij}^{AA} + J_{ij}^{BB} - 2J_{ij}^{AB} \right) \quad \text{Magnetic}$$

+ ({Moment change effects})

Effect of "exchange splitting" on electronic structure $Ni_{75}Fe_{25}$ f.c.c. ferromagnetic & paramagnetic

$\{V_i^{ext}\}$ also induces $\{\delta\mu_i\}$

$\rightarrow \dfrac{\partial \mu_i}{\partial V_j^{ext}}$

occupation of site i

$$\gamma'(\vec{q}) = \beta \sum_j \langle \varrho_i \mu_j \rangle e^{i\vec{q}\cdot\vec{R}_{ij}}$$

moment on site j

Magnetic - compositional correlation function

$$= \alpha(\vec{q}) \cdot \gamma(\vec{q})$$

Compositional correlations

Apply external magnetic field $\{h_i^{ext}\}$

$\longrightarrow \{\delta c_i\}$ as well as $\{\delta\mu_i\}$

Magnetic correlations

$$\chi(\vec{q}) = \beta \sum_j \langle \mu_i \mu_j \rangle e^{i\vec{q}\cdot\vec{R}_{ij}}$$

$$= \bar{\chi}(\vec{q}) + \left(\gamma(\vec{q})\right)^2 \alpha(\vec{q})$$

Compositional correlations

(+ $\gamma'(\vec{q})$ again)

11.

Polarised neutron scattering:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\epsilon=\pm 1} = \left(\frac{d\sigma}{d\Omega}\right)_{Nuclear}^{\alpha(\vec{q})} + \epsilon\left(\frac{d\sigma}{d\Omega}\right)_{Nuc-Mag.}^{\alpha(\vec{q})\gamma(\vec{q})}$$

Polarisation

$$+\left(\frac{d\sigma}{d\Omega}\right)_{Mag.} \leftarrow \chi(\vec{q})$$

Return to consider $\gamma'(\vec{q})$ magnetic–compositional "cross" correlations.

$$\gamma'(\vec{q}) = \alpha(\vec{q})\,\gamma(\vec{q})$$

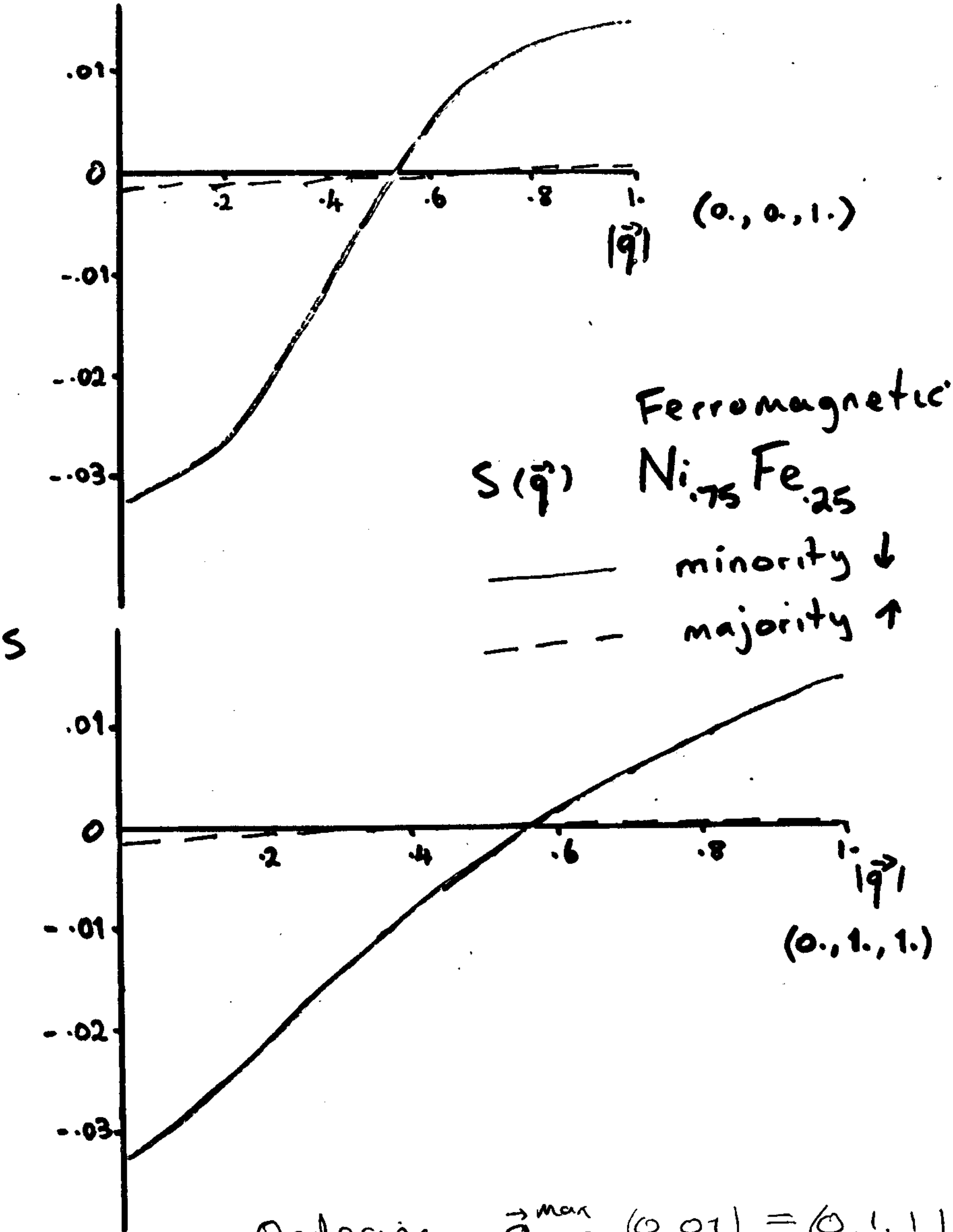
$$\gamma(\vec{q}) = \cancel{(\bar{\mu}_A - \bar{\mu}_B)} + c\,\gamma_A(\vec{q}) + (1-c)\,\gamma_B(\vec{q})$$

$$\gamma_{A(B)}(\vec{q}) = \sum_j \gamma_{A(B)}^{ij}\, e^{i\vec{q}\cdot R_{ij}}$$

$$= \sum_j \frac{\partial \mu_i^{A(B)}}{\partial c_j}\, e^{i\vec{q}\cdot\vec{R}_{ij}}$$

Dependence of magnetic moments on chemical environment.

7.19



Ordering $\vec{q}^{\,max} = (0,0,1) \equiv (0,1.,1.)$

$L1_2$ structure

J.W. Cable, H.R. Child + Y. Nakai

reported on diffuse unpolarised neutron scattering measurements on $Fe_{86.5}V_{13.5}$ single crystal (Physica 156 & 157, 50 (1989))

$\rightarrow \frac{d\sigma}{d\omega}^{Nuc.}$ Meaning of $\gamma_{A(b)}$ [1,0,0], [1,1,0] [1,1,1]

$= \frac{d\sigma}{d\omega}^{incoherent} + \frac{d\sigma}{d\omega}^{mult.} + |\Delta b|^2 \cancel{\alpha(q)}$

We compare ~~with~~ their measurements using same estimates of $\left(\frac{d\sigma}{d\omega} + \frac{d\sigma}{d\omega}^{mult.}\right)$ and Δb as they used.

Same 'quench' ~~temperature~~ used.

No adjustable parameters.

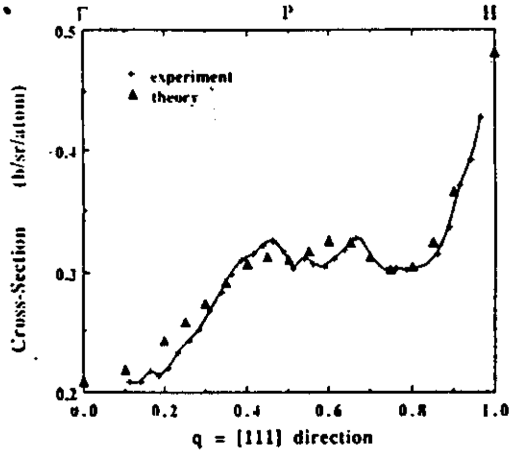
9.

$\boxed{Fe_{87}V_{13}}$

$\left(\frac{d\sigma}{d\Omega}\right)^{Nuc.}$

+++ expt.

▲▲▲ theory

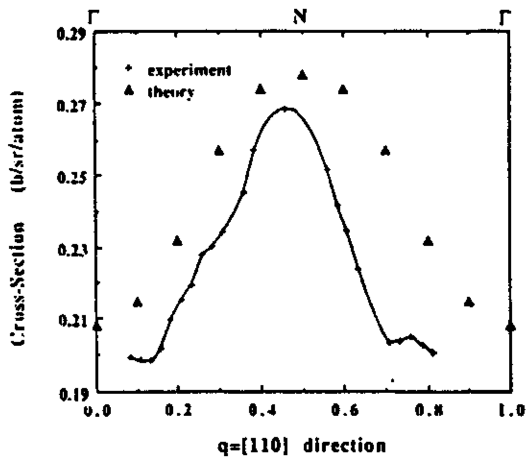


1a

$\vec{q}$

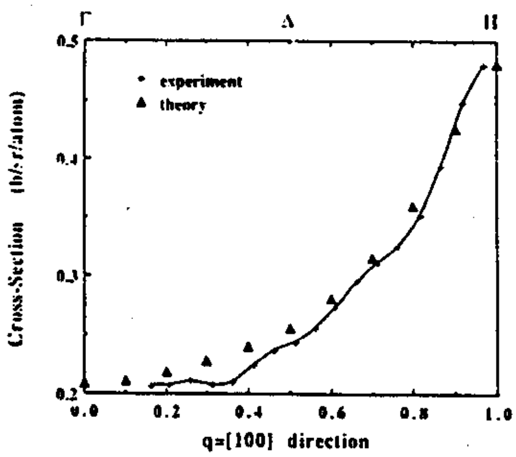
[1, 1, 1]

NB. different scale



[1, 1, 0]

1b



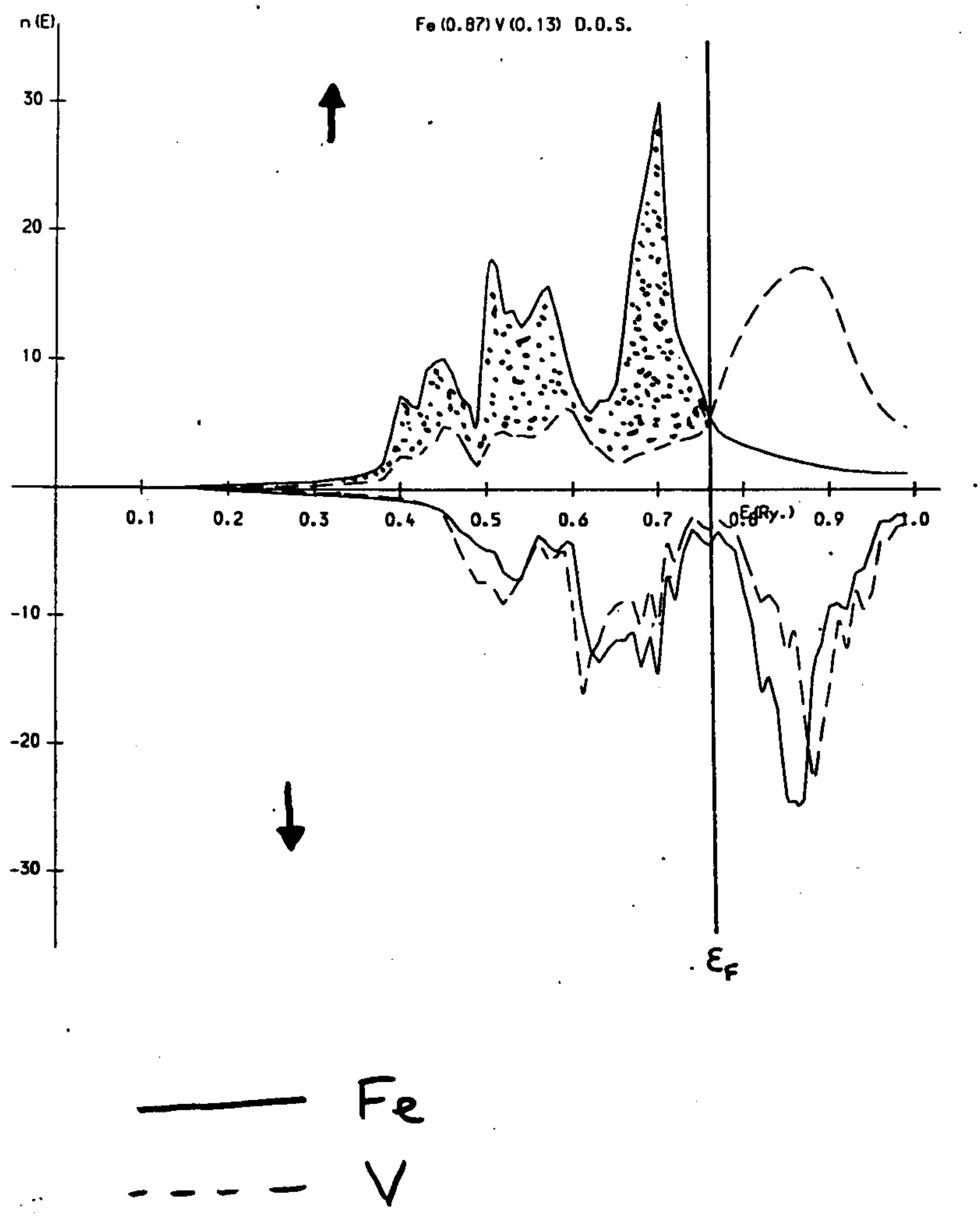
[1, 0, 0]

1c

Staunton et al. (1990) PRL.

Figure 1. Comparison of the calculated and experimental nuclear cross-sections for BCC $Fe_{0.865}V_{0.135}$ along the a) [111], b) [110], and c) [100] wave-vector directions.

15.
14.


n(E)
Fe(0.87)V(0.13) D.O.S.
30
20
10
-10
-20
-30
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
E(Ry.)
ε_F
Fe
V

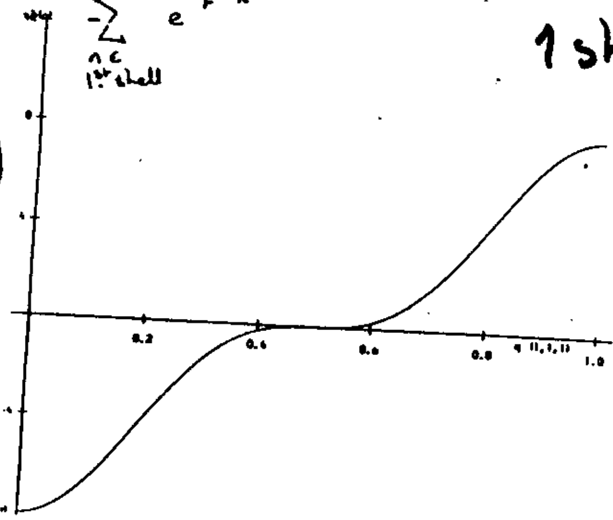
16.
15.

$$\alpha(q) = \frac{\beta c(1-c)}{\left(1-\beta c(1-c) S(q)\right)}$$

$$S(q) = \sum_{\substack{\text{shells} \\ n}} \sum_{i \in n} e^{i q R_i^n} S_n$$

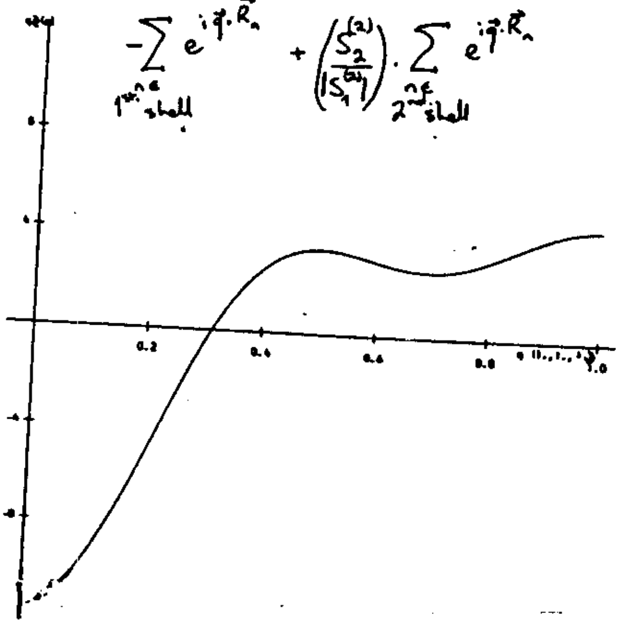
$$-\sum_{\substack{n.c. \\ 1^{st}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n}$$

1 shell



$$-\sum_{\substack{n.c. \\ 1^{st}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_2^{(2)}}{|S_1^{(2)}|}\right) \cdot \sum_{\substack{n.c. \\ 2^{nd}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n}$$

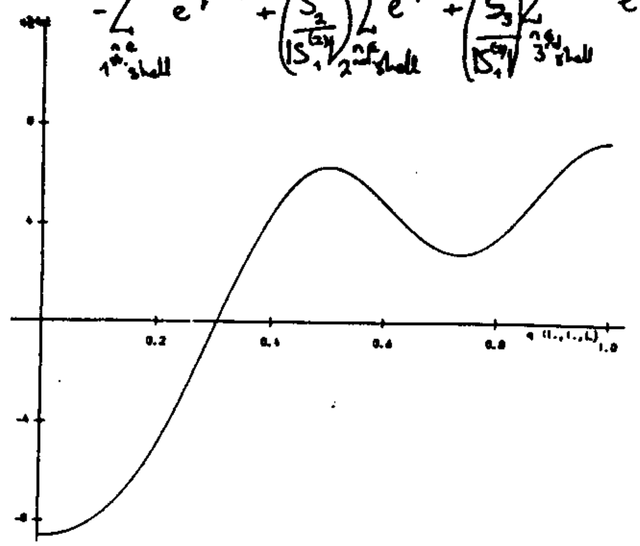
[1,1,1]



2shells

$$-\sum_{\substack{n.c. \\ 1^{st}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_2^{(3)}}{|S_1^{(3)}|}\right) \sum_{\substack{n.c. \\ 2^{nd}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n} + \left(\frac{S_3^{(3)}}{|S_1^{(3)}|}\right) \sum_{\substack{n.c. \\ 3^{rd}\,\text{shell}}} e^{i\vec{q}\cdot\vec{R}_n}$$

3 shells



17.
16.

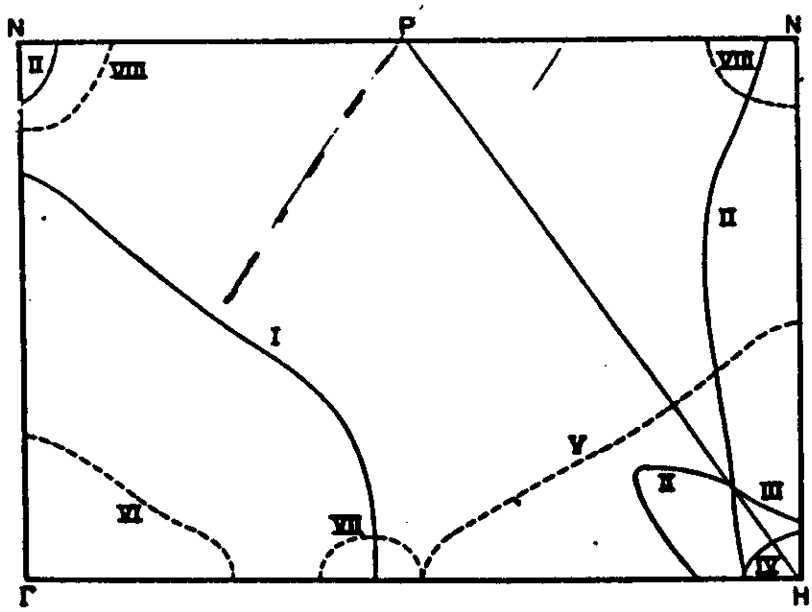


FIG. 4. Cross section of the Fermi surface in a (110) plane.

Fe b.cc. Fermi surface

(Callaway + Wang)

——— majority

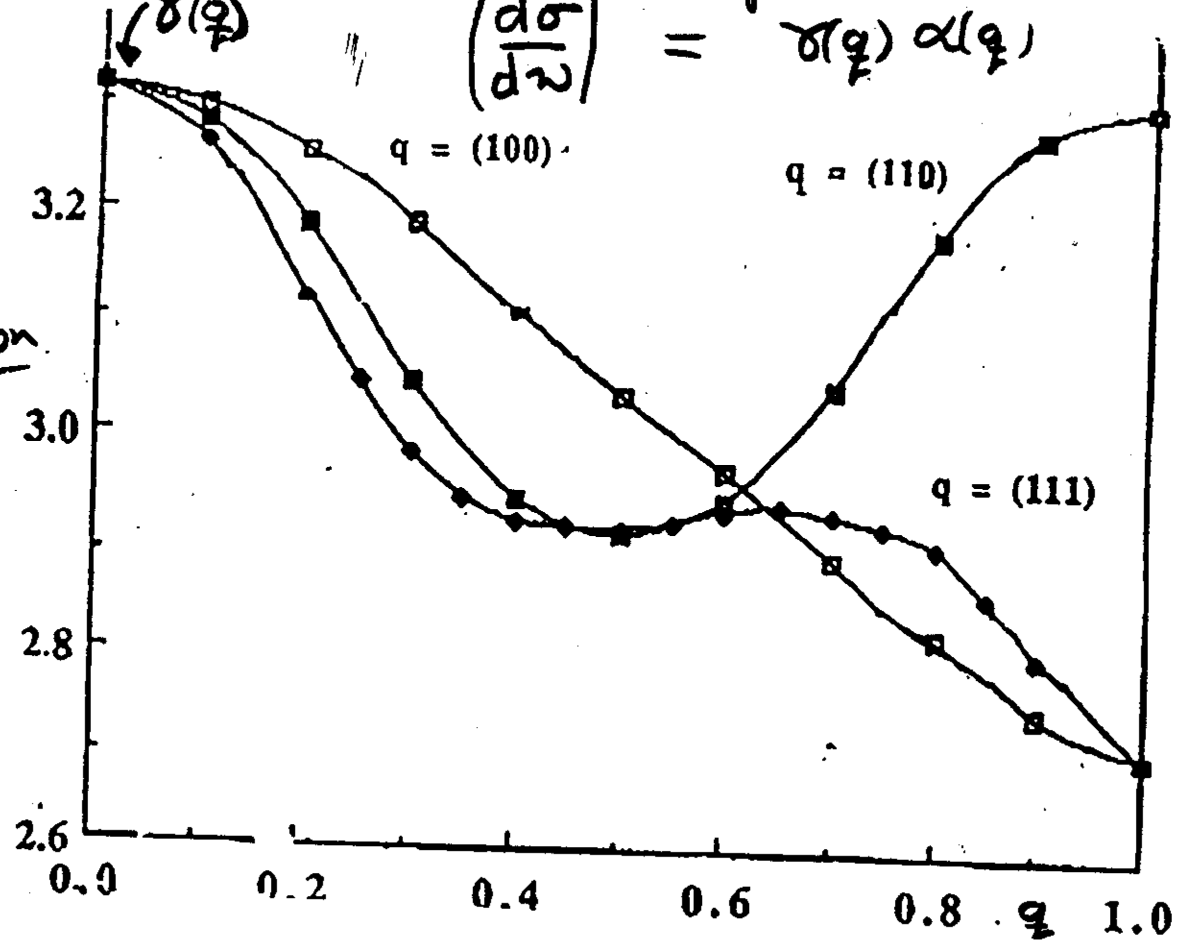
······· minority

18.
17.

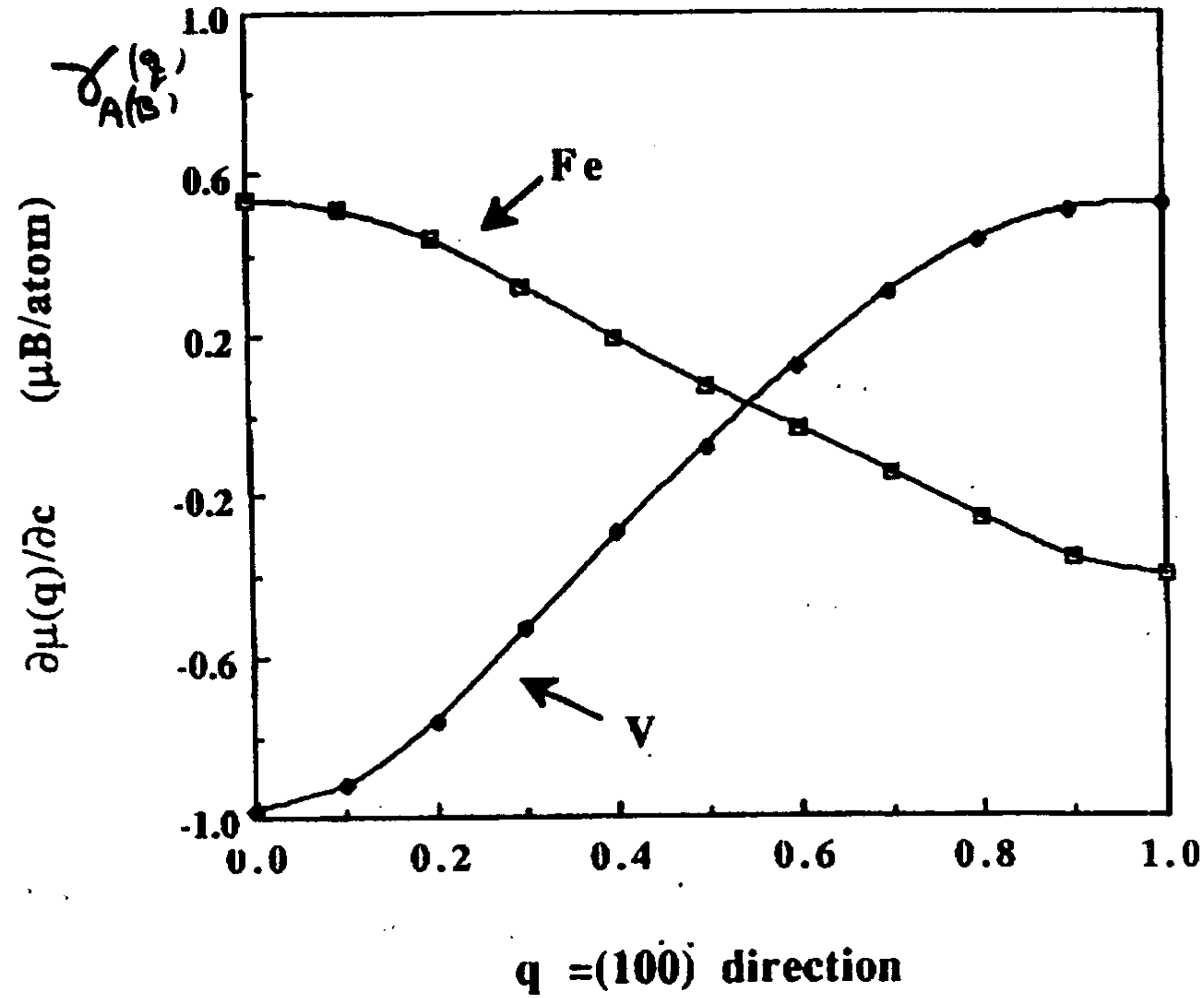
Fe-13.5%V Gammas

$$\left(\frac{d\sigma}{d\omega}\right)^{\text{Nuc. Mag}} = \gamma(q)\,\alpha(q)$$

<u>Prediction</u>



Fe-13.5% V



$$\gamma(q) = (\mu_A - \mu_B) + c\,\gamma_A(q) + (1-c)\,\gamma_B(q)$$

Real space

$\gamma_{ij}^{A(B)}(1-c)$ .... change in moment of an $A^{(B)}$ atom in disordered alloy on site i if site j now is <u>definitely</u> occupied by A atom

$\dfrac{\partial \mu_i^{A(B)}}{\partial c_j}$

$\gamma_{ij}^{A(B)}(-c)$ --- site j with B atom.

Use to study effects on magnetic properties from various chemical S.R.O ---e.g $\bar{\mu}$ of Ni_3Fe 5% greater than that in disordered $Ni_{75}Fe_{25}$

Modulated alloys ?

Theoretical alloy design?

- Theory of compositional and magnetic correlations in alloys

  → alloy design, modulated alloys, magnetic multilayers

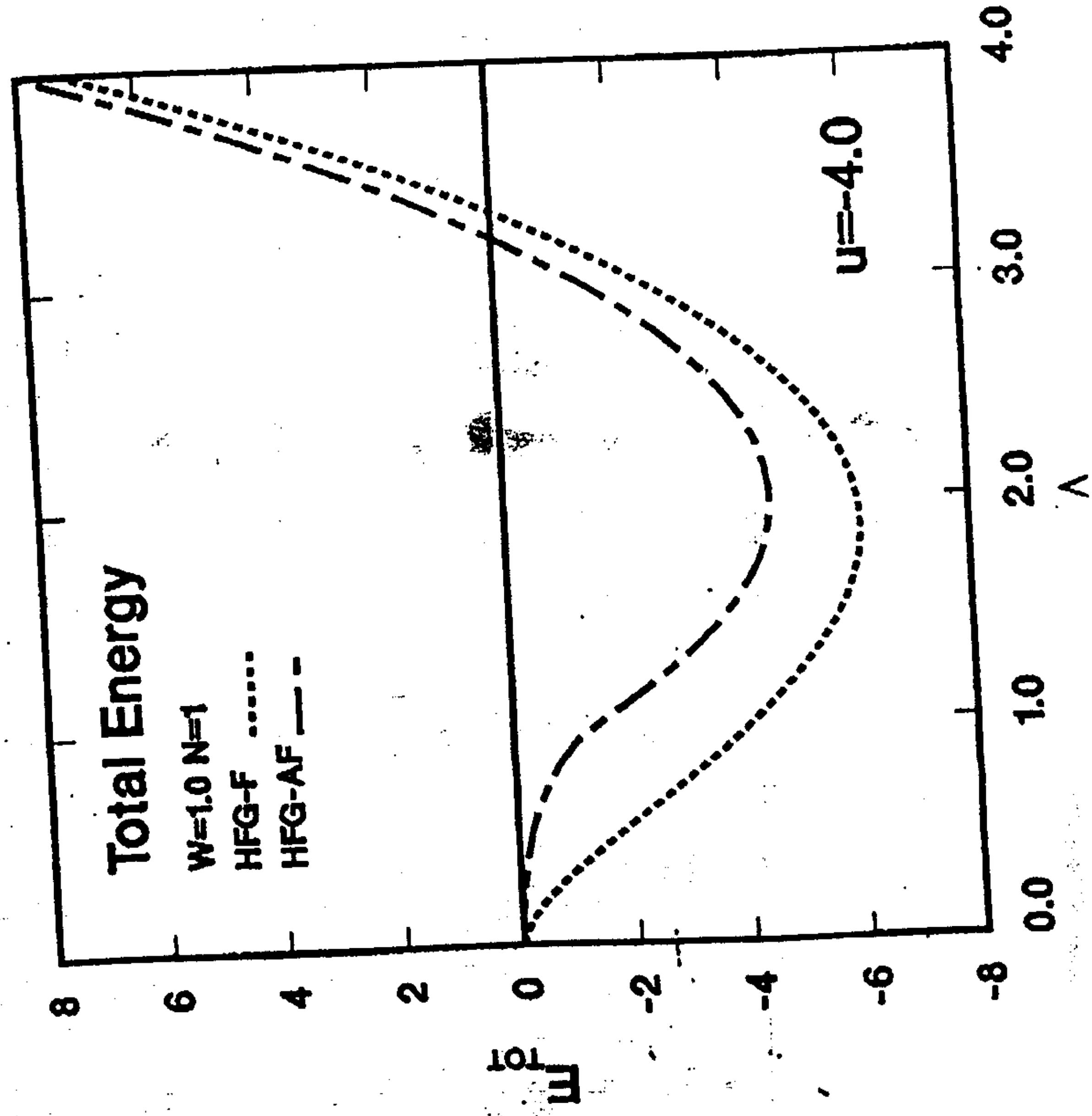
- Test: $Fe_{86.5}V_{13.5}$ single crystal
  $Ni_{75}Fe_{25}$

  Comparison in q-space.

- Excellent comparison with unpolarised neutron scattering data. $\alpha(q)$ Compositional correlations

- Prediction for polarised experiment $\gamma(q)$ Magnetic response to compositional environment

- Future fruitful collaboration between theory and experiment


Total Energy
W=1.0 N=1
HFG-F
HFG-AF
u=-4.0
E_{TOT}
Λ
0.0
1.0
2.0
3.0
4.0
8
6
4
2
0
-2
-4
-6
-8

1.

Itinerant Magnetism at finite temperatures

JBS , B. L. Gyorffy

Nature of paramagnetic state of ferromagnetic metals ??

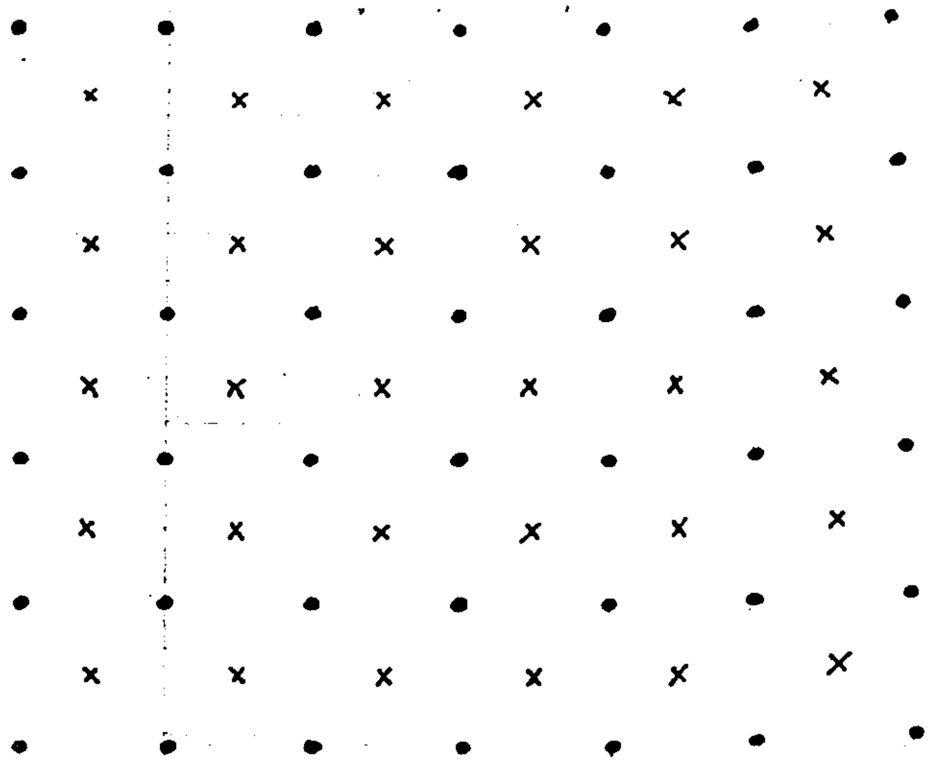


Fig 9

Stoner-Wohlfarth-Hubbard Model

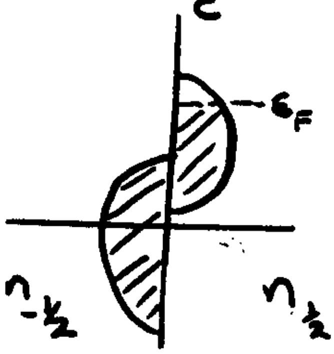
$$H = \sum_{\substack{ij\\ \sigma}} (\varepsilon_0 \delta_{ij} + t_{ij})\, a^+_{i\sigma} a_{j\sigma} + \tfrac{1}{2} I \sum_{i\sigma} a^+_{i\sigma} a_{i\sigma} a^+_{i-\sigma} a_{i-\sigma}$$

$$a^+_{i\sigma} a_{i\sigma} a^+_{i-\sigma} a_{i-\sigma} \Rightarrow a^+_{i\sigma} a_{i\sigma} \underbrace{\langle a^+_{i-\sigma} a_{i-\sigma} \rangle}_{\bar{n}_{i,-\sigma}}$$

$$\bar{n}_i = \bar{n}_{i\frac{1}{2}} + \bar{n}_{i-\frac{1}{2}}\,, \quad \bar{\mu}_i = \bar{n}_{i\frac{1}{2}} - \bar{n}_{i-\frac{1}{2}} = \tfrac{1}{2}\bar{n}_i - \tfrac{1}{2}\bar{\mu}_i \sigma$$

$$H_{HF} = \sum_{ij\sigma} \left[(\varepsilon_0 + \tfrac{1}{2} I \bar{n}_i - \tfrac{1}{2} I \bar{\mu}_i \sigma)\delta_{ij} + t_{ij} \right] a^+_{i\sigma} a_{j\sigma}$$

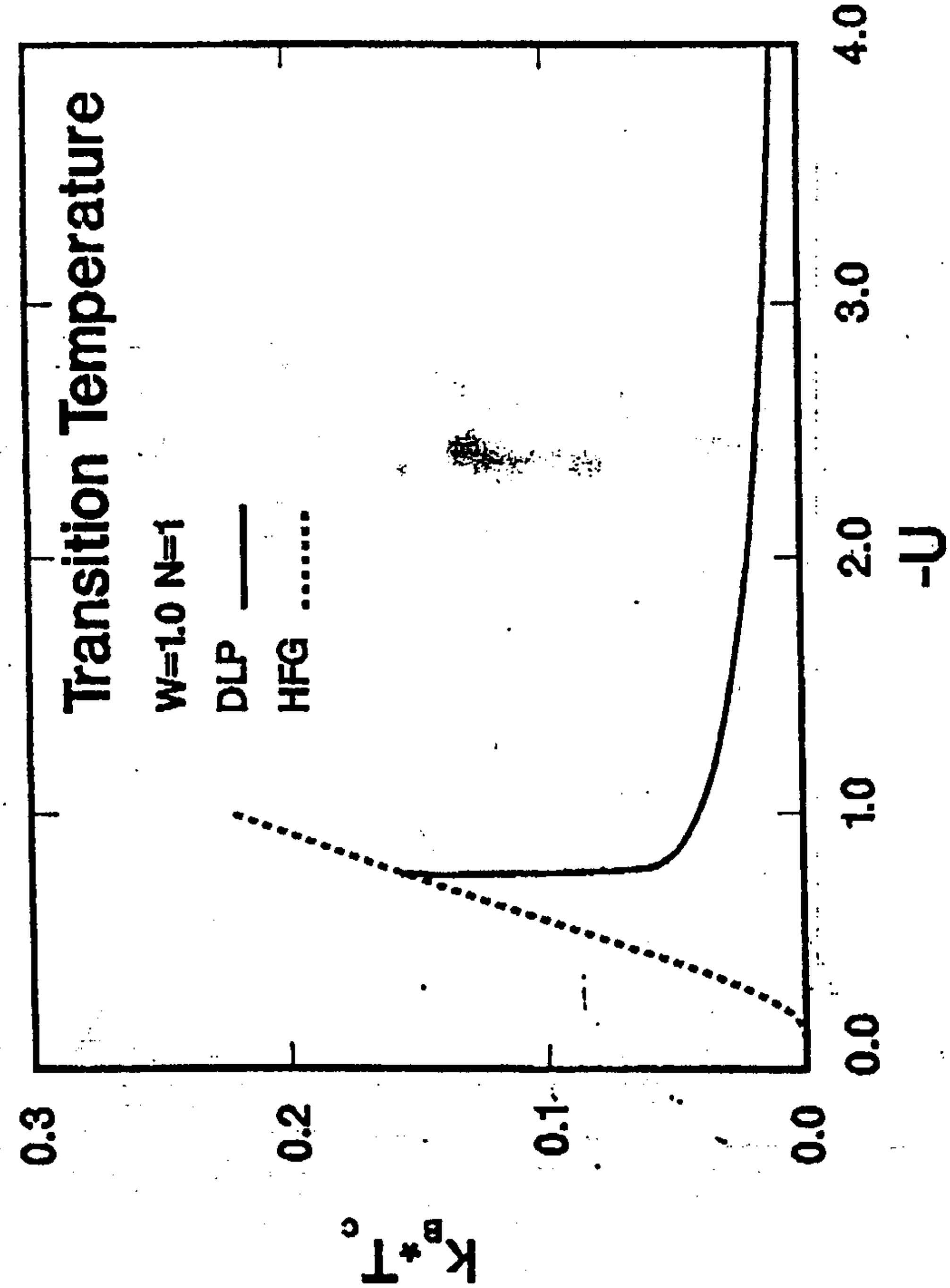
(Spin polarized band theory)



Modern spin-polarised band theory with basis in Spin Density Functional Theory is successful in describing itinerant magnetic properties in transition metals at $T = 0$.

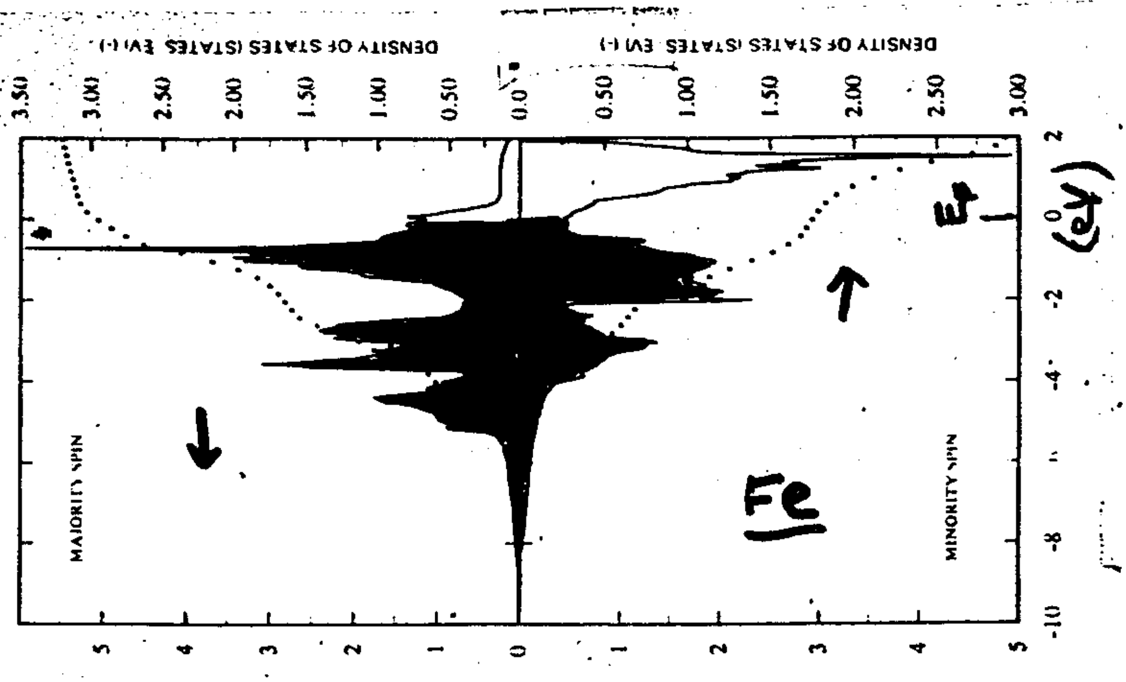
e.g. Correct exchange splitting (? !)
Magnetic moment (non-integer)
Cohesive energy
Magnetic instability etc.

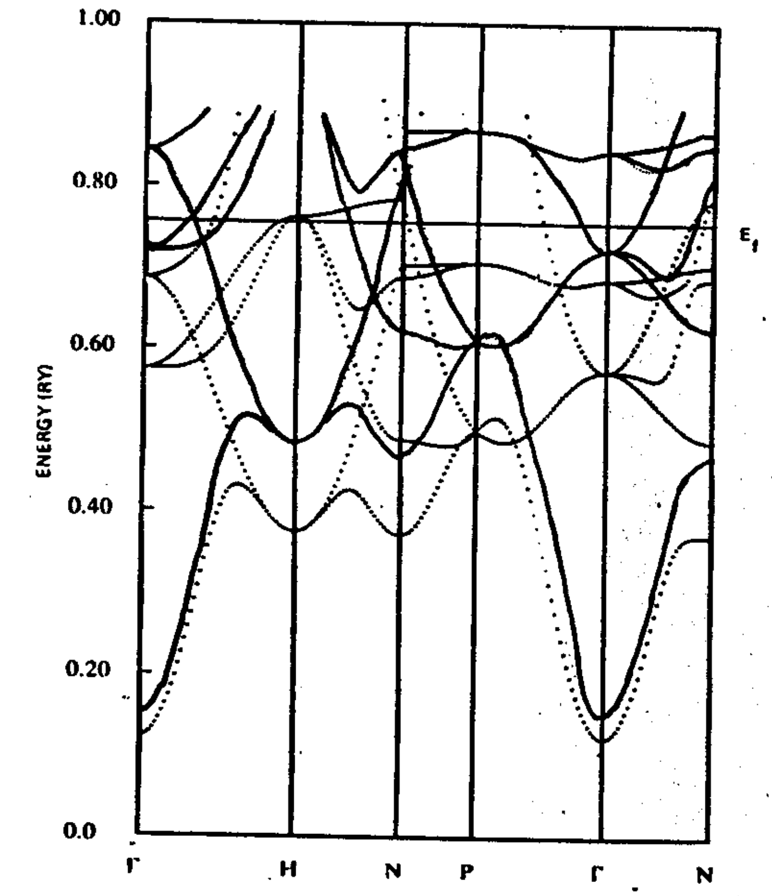
Fig 8


Transition Temperature
W=1.0 N=1
DLP
HFG
0.0
1.0
2.0
3.0
4.0
-U
0.0
0.1
0.2
0.3
K_B*T_c

3.

IIe



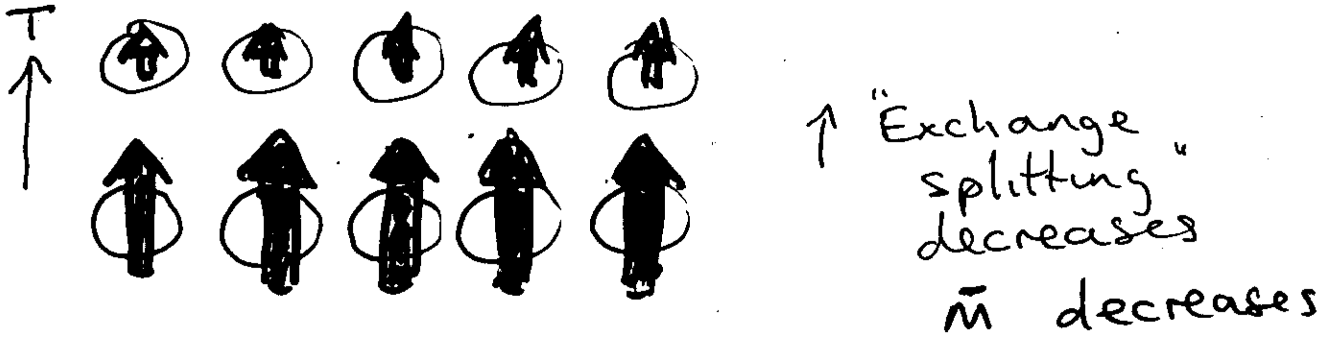


Dark points for majority spin, light points for minority spin.

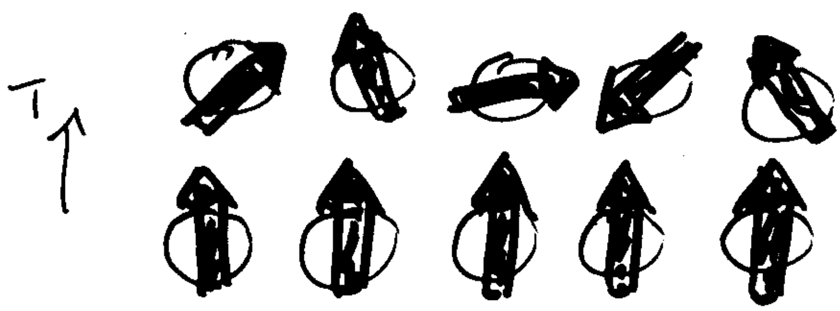
$$\left\{-\nabla^2 + v^{eff}[n,\vec{m}]\,\underset{\sim}{1} + \underset{\sim}{\sigma}\cdot B^{eff}[n,\vec{m}]\right\} \underset{\sim}{\phi}_i(\vec{r},\varepsilon) = \varepsilon_i\, \underset{\sim}{\phi}_i(\vec{r},\varepsilon)$$

Kohn-Sham single particle equation

Straightforward generalisation to finite T fails. T_c too high
no "local moments"
no Curie Weiss law



Improved theory:



Picture: Stoner - Wohlfarth "spin polarised band theory + thermal fluctuations of local moments

Paramagnetic state:

- Itinerant electrons + spin fluctuations

- Time scale separation

spin fluctuation orientations vary slowly on scale of electron hopping time

i.e. $\hbar\omega_{sf} \ll W$

but $k_B T < \hbar\omega_{sf}$ (thermal averages)

"Local Moment" picture

- + Long wavelength, low frequency fluctuations $k_B T > \hbar\omega_{slow}$

"Weak itinerant" magnets

- 'First principles' description SDF theory. Electronic structure basis

Outline:

- Recap of Disordered Local Moment picture.
  Mean field theory

- Improvements via Onsager Cavity field construction

- Deals with good 'Local Moment' systems and (high T, static limit)
  Weak itinerant magnets

- Results Fe, Ni.

8.

Time scale separation

- $\vec{M}_{\tau,i} = \mu_i\{\hat{e}_k\}\,\hat{e}_i$ **Local moment**

$\{\hat{e}_k\}$ orientations slowly vary

$\{\mu_k\}$ fluctuate rapidly

Generalisation of Spin Density Functional theory $\Omega\{\hat{e}_i\}, \mu_k\{\hat{e}_i\}$

Long time averages

$$P\{\hat{e}_i\} = \frac{e^{-\beta\Omega\{\hat{e}_i\}}}{\prod_i \int d\hat{e}_i \exp -\beta\Omega\{\hat{e}_k\}}$$

$$F = -k_B T \ln Z$$

Entropy of orientational fluctuations + creation of electron-hole pairs (Stoner excitations) a

Mean field theory

$\rightarrow \quad \langle \Omega\{\hat{e}_k\}\rangle_{\hat{e}_i}, \ \langle \mu_i\{\hat{e}_k\}\rangle_{\hat{e}_i}$

SCF-KKR-CPA

e.g. Paramagnetic state

$\bar{\mu} = 1.9\,\mu_B$ Fe b.c.c.

$T_c^{MF} \sim 1300K$ ($T_{expt} \sim 1040K$)

($\checkmark$)

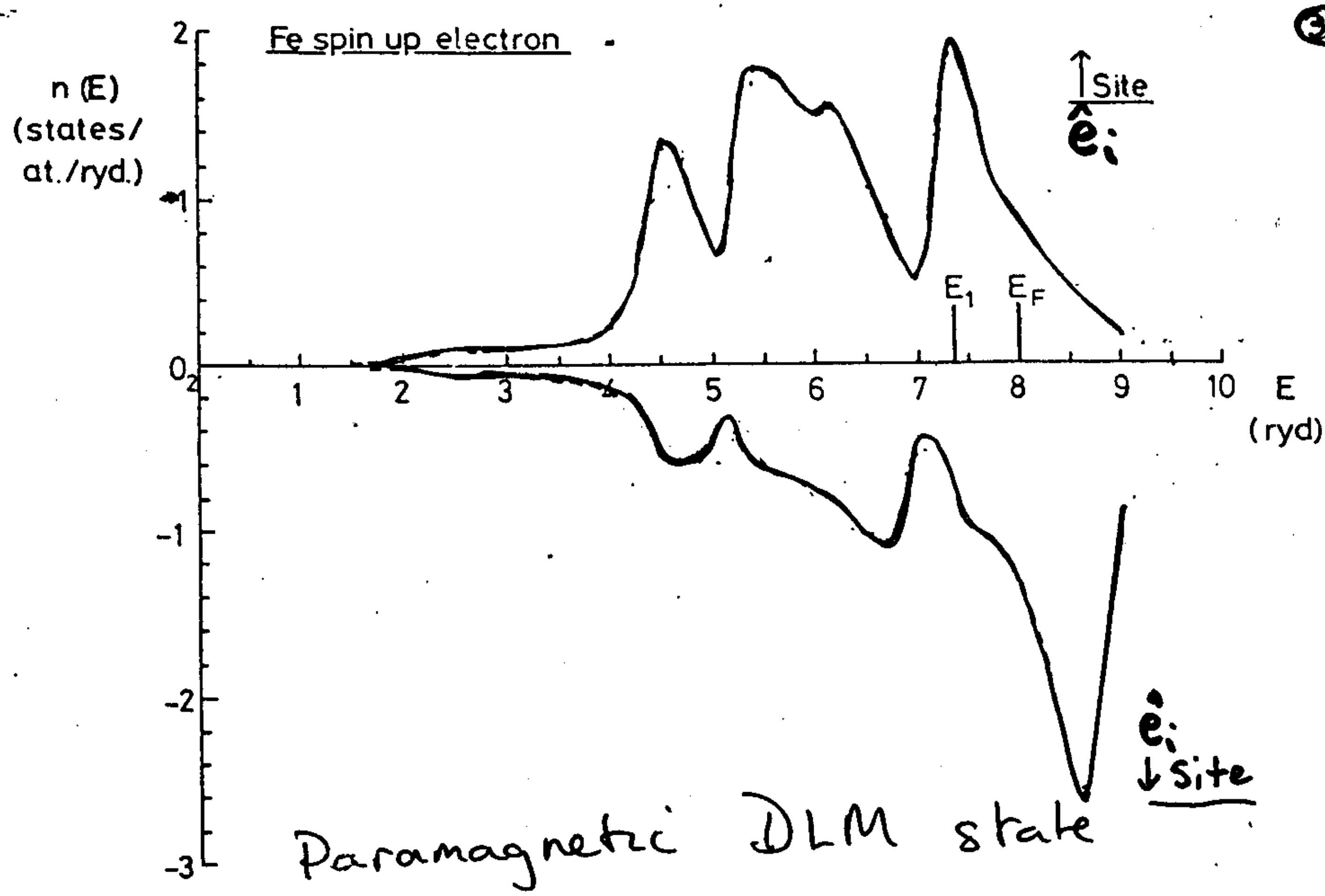
$$\vec{M} = \int \underbrace{P_i(\hat{e}_i)}_{1/4\pi} \underbrace{\mu_i(\hat{e}_i)}_{\bar{\mu}} \hat{e}_i = 0$$

$\bar{\mu} = 0$ Ni f.c.c.

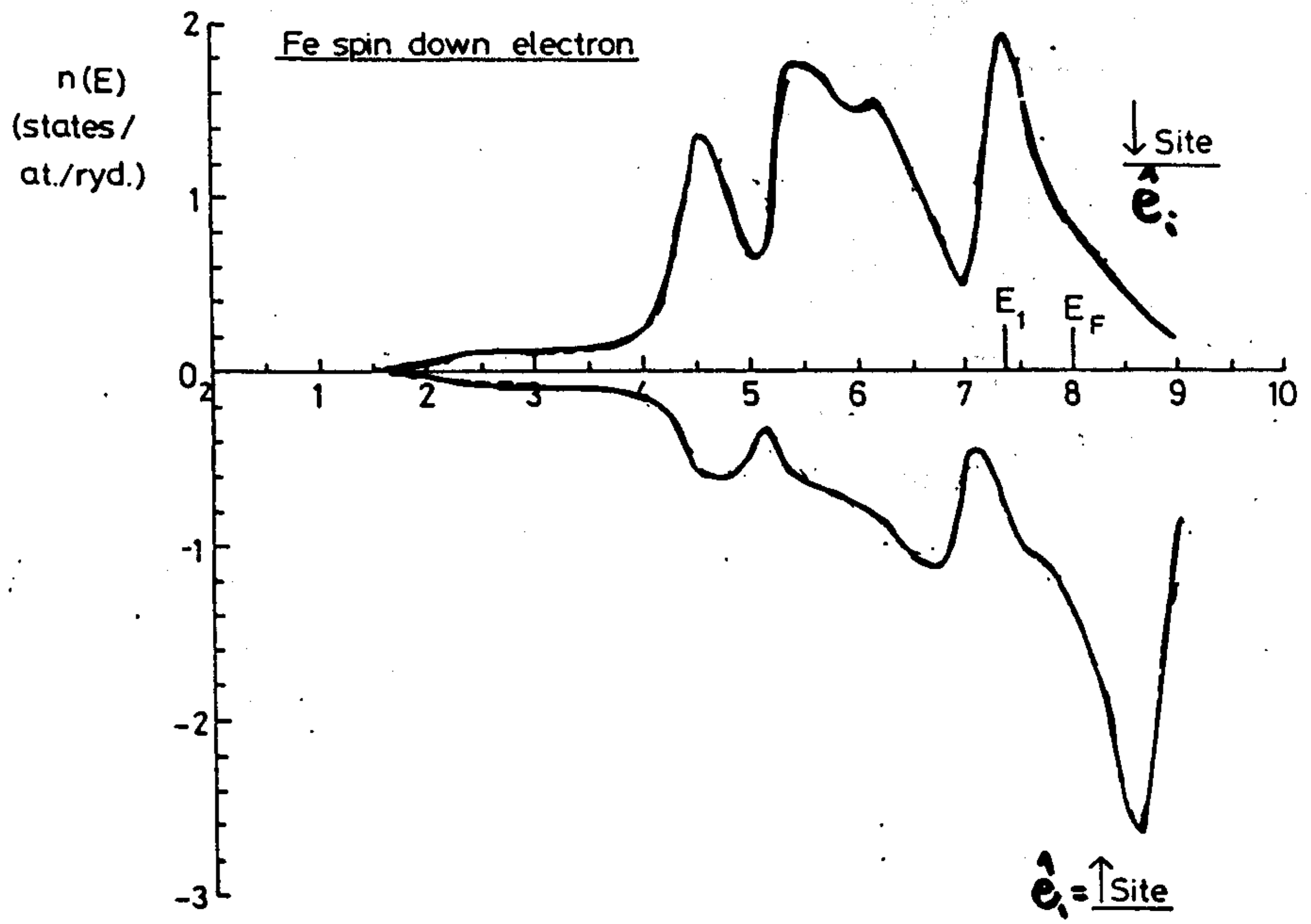
$T_c^{MF} = T_s \sim 3000K$ (660K)

X

+ Onsager Cavity field.



Paramagnetic DLM state



6.

Classical Heisenberg model

$$\hat{H} = -\sum_{ij} J_{ij}\, \hat{e}_i \cdot \hat{e}_j \, .$$

$\{\vec{h}_j\}$ external mag. field on paramagnetic system

$$\vec{m}_i = \langle \hat{e}_i \rangle$$

$$\vec{m}_i \doteq \beta/3 \Big(\underbrace{\sum_j J_{ij}\, \vec{m}_j + \vec{h}_i}_{\text{Weiss } \vec{h}^w_i} \Big) \qquad \text{MFA}$$

$$\chi(\vec{q}) = \frac{\beta/3}{(1 - \beta/3\, J(\vec{q}))}$$

$$\vec{m}_i \doteq \beta/3 \Big(\underbrace{\sum_j J_{ij}(\vec{m}_j - \delta\vec{m}_j^{(i)}) + \vec{h}_i}_{\text{Cavity field } \vec{h}^c_i} \Big)$$

$$\delta\vec{m}_j^{(i)} = \tilde{\chi}_{ji}\, \vec{h}^c_i$$

$$\vec{m}_i = \tilde{\chi}_{ii}\, \vec{h}^c_i$$

$$\delta m_j^{(i)} = \tilde{\chi}_{ji}\, \tilde{\chi}_{ii}^{-1}\, \vec{m}_i$$

7.

$$\boxed{\begin{aligned} \chi(\vec{q}) &= \beta/3 \left[(J(\vec{q}) - \lambda)\chi(\vec{q}) + 1 \right] \\ \lambda &= \chi_{ii}^{-1} \int d\vec{q}\, J(\vec{q})\, \chi(\vec{q}) \end{aligned}}$$

$$\chi_{ii} = \beta/3 \,, \quad \langle \hat{e}_i^2 \rangle = \frac{3}{\beta} \chi_{ii}$$

Fluctuation Dissipation

Spherical approximation

Analog for DLM itinerant electron system?

Apply $\{\vec{h}_\ell\}$ to paramagnetic system

$$\{\delta P_\ell(\hat{e}_\ell)\}$$

$$\{\delta\mu_\ell(\hat{e}_\ell)\}$$

Induced magnetisation

$$\vec{M}_i = \int d\hat{e}_i\, \mu_i(\hat{e}_i)\, \hat{e}_i\, P(\hat{e}_i)$$

$$\simeq \frac{1}{4\pi}\int d\hat{e}_i\, \delta\mu_i(\hat{e}_i)\hat{e}_i + \bar{\mu}\int d\hat{e}_i\, \hat{e}_i\, \delta P_i(\hat{e}_i)$$

$$= \vec{\mu}_i + \bar{\mu}\,\vec{m}_i$$

$$\chi_{ij} = \chi^{\mu}_{ij} + \chi^{m}_{ij}$$ Susceptibili[ty]

Moments magnitude response **Orientational response**

$$\bar{\mu}\vec{m}_i = \frac{\beta}{3}\sum_{l\neq i}\left(\bar{\mu}S^{mm}_{il}(\vec{m}_l - \delta\vec{m}^{(i)}_l) + \bar{\mu}S^{m\mu}_{il}(\vec{\mu}_l - \delta\vec{\mu}^{(i)}_l)\right)$$
$$+ \beta/3 \sum_l \left(\bar{\mu}^2\delta_{il} + \bar{\mu}\,\Xi_{il}(1-\delta_{il})\right)\vec{h}_l$$

$$\vec{\mu}_i = \sum_{l\neq i}\gamma^{\mu m}_{il}(\vec{m}_l - \delta\vec{m}^{(i)}_l)$$
$$+ \sum_l \gamma^{\mu\mu}_{il}(\vec{\mu}_l - \delta\vec{\mu}^{(i)}_l) + \sum_l \chi^{\circ}_{il}\vec{h}_l$$

S^{mm}_{il}, $S^{m\mu}_{il}$, Ξ_{il}, $\gamma^{\mu m}_{il}$, $\gamma^{\mu\mu}_{il}$, χ°_{il}

.... SCF-KKR-CPA.

$$\bar{\mu}\,\delta\vec{m}^{(i)}_l = \tilde{\chi}^m_{li}\vec{h}^c_i = \tilde{\chi}^m_{li}\hat{\chi}^{-1}_{ii}\vec{M}_i$$
$$\delta\vec{\mu}^{(i)}_l = \tilde{\chi}^\mu_{li}\vec{h}^c_i = \hat{\chi}^\mu_{li}\tilde{\chi}^{-1}_{ii}\vec{M}_i$$

Closed set of equations ....

$$\chi^{m}(\vec{q}) = \frac{\beta}{3}\Big(S^{mm}(\vec{q})\,\chi^{m}(\vec{q}) + \mu\, S^{m\mu}(\vec{q})\,\chi^{\mu}(\vec{q})$$
$$- \Lambda_1\big(\chi^{m}(\vec{q}) + \chi^{\mu}(\vec{q})\big)$$
$$+ \bar{\mu}^2 + \bar{\mu}\,\Xi(\vec{q})\Big)$$

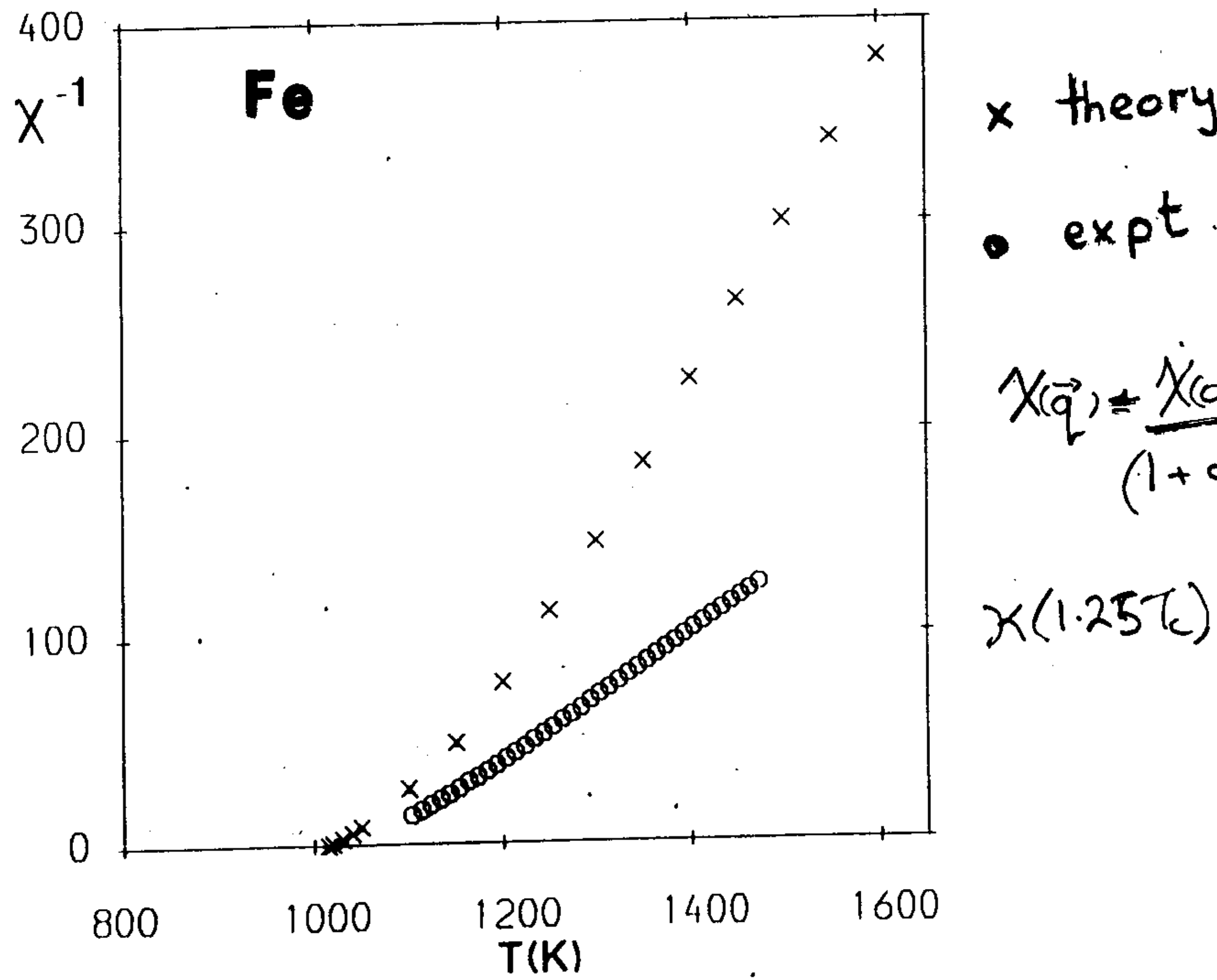
$$\chi^{\mu}(\vec{q}) = \gamma^{\mu m}(\vec{q})\,\chi^{m}(\vec{q}) + \gamma^{\mu\mu}(\vec{q})\,\chi^{\mu}(\vec{q})$$
$$- \Lambda_2\big(\chi^{m}(\vec{q}) + \chi^{\mu}(\vec{q})\big)$$
$$+ \chi^{0}(\vec{q})$$

$$\Lambda_1 = \chi_{ii}^{-1}\int d\vec{q}\,\big(S^{mm}(\vec{q})\,\chi^{m}(\vec{q}) + \bar{\mu}\, S^{m\mu}(\vec{q})\,\chi^{\mu}(\vec{q})\big)$$

$$\Lambda_2 = \chi_{ii}^{-1}\int d\vec{q}\,\big(\gamma^{\mu m}(\vec{q})\,\chi^{m}(\vec{q}) + \gamma^{\mu\mu}(\vec{q})\,\chi^{\mu}(\vec{q})\big)$$

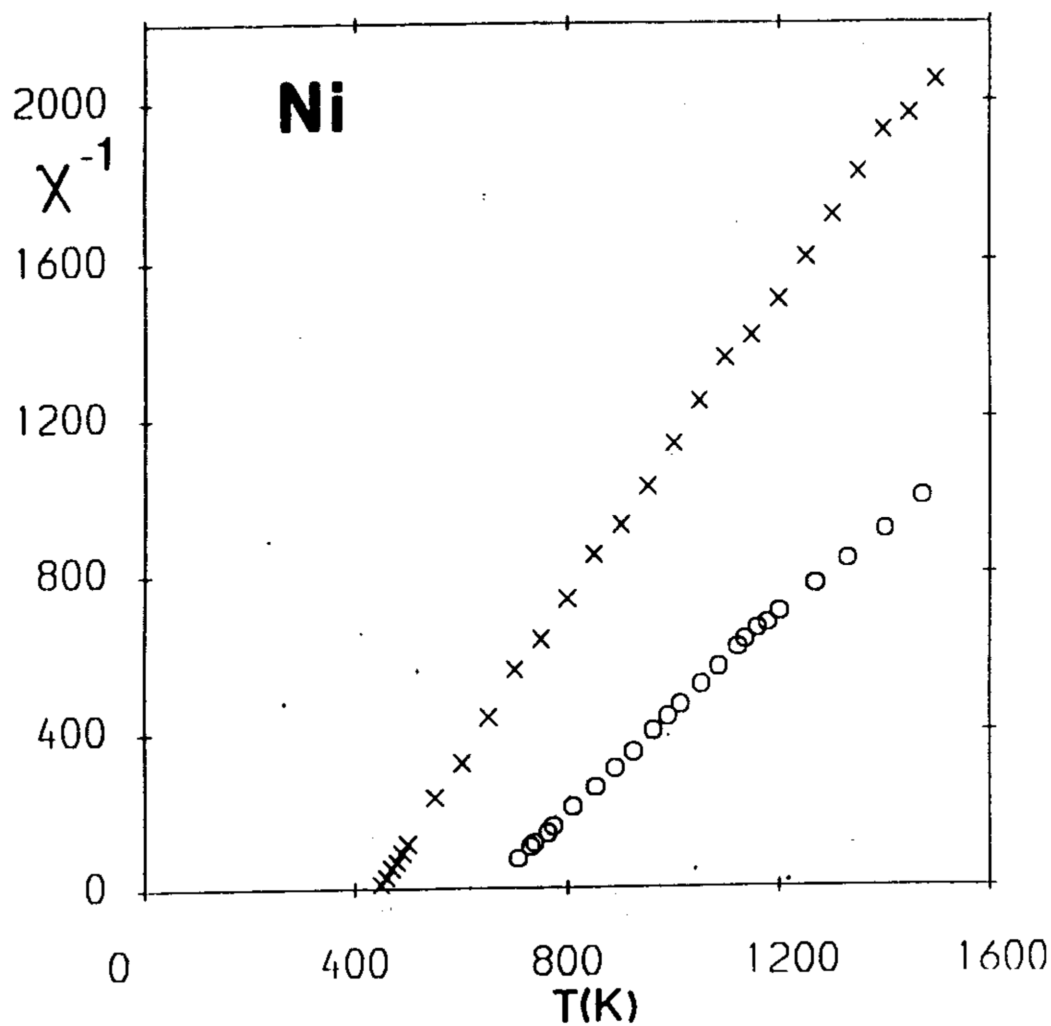
$$\langle M_i^2\rangle = \bar{\mu}^2 + \frac{3\chi_{ii}^{0}}{\beta}$$

Staunton & Gyorffy (1992) PRL 69, 371



$$\chi(\vec{q}) \doteq \frac{\chi(0)}{(1+q^2/\kappa^2)}$$

$$\kappa(1.25T_c) \sim .37\mathring{A}^{-1}$$
$$(.4\mathring{A}^{-1}\text{ neutron})$$



× theory

○ expt.

$\chi_{th} \sim .28\,\mathring{A}^{-1}$ $\quad 1.25\,T_c$

$\chi_{ex} \sim .22\,\mathring{A}^{-1}$

$$\chi(\vec{q}) = \frac{\chi_0(\vec{q})}{\left(1 - I\chi_0(\vec{q}) + \Lambda\right)}$$

$$\Lambda = (\chi^0_{ii})^{-1} \int d\vec{q}\, I \chi_0(\vec{q})\, \chi(\vec{q})$$

6.

Compositional and Magnetic Ordering in Alloys + M. F. Ling (Warwick)

Preliminary calculations NiFe, CuMn

Starting point: High T compositionally disordered, paramagnetic state.

Compare Stoner and DLM models of paramagnetic state.

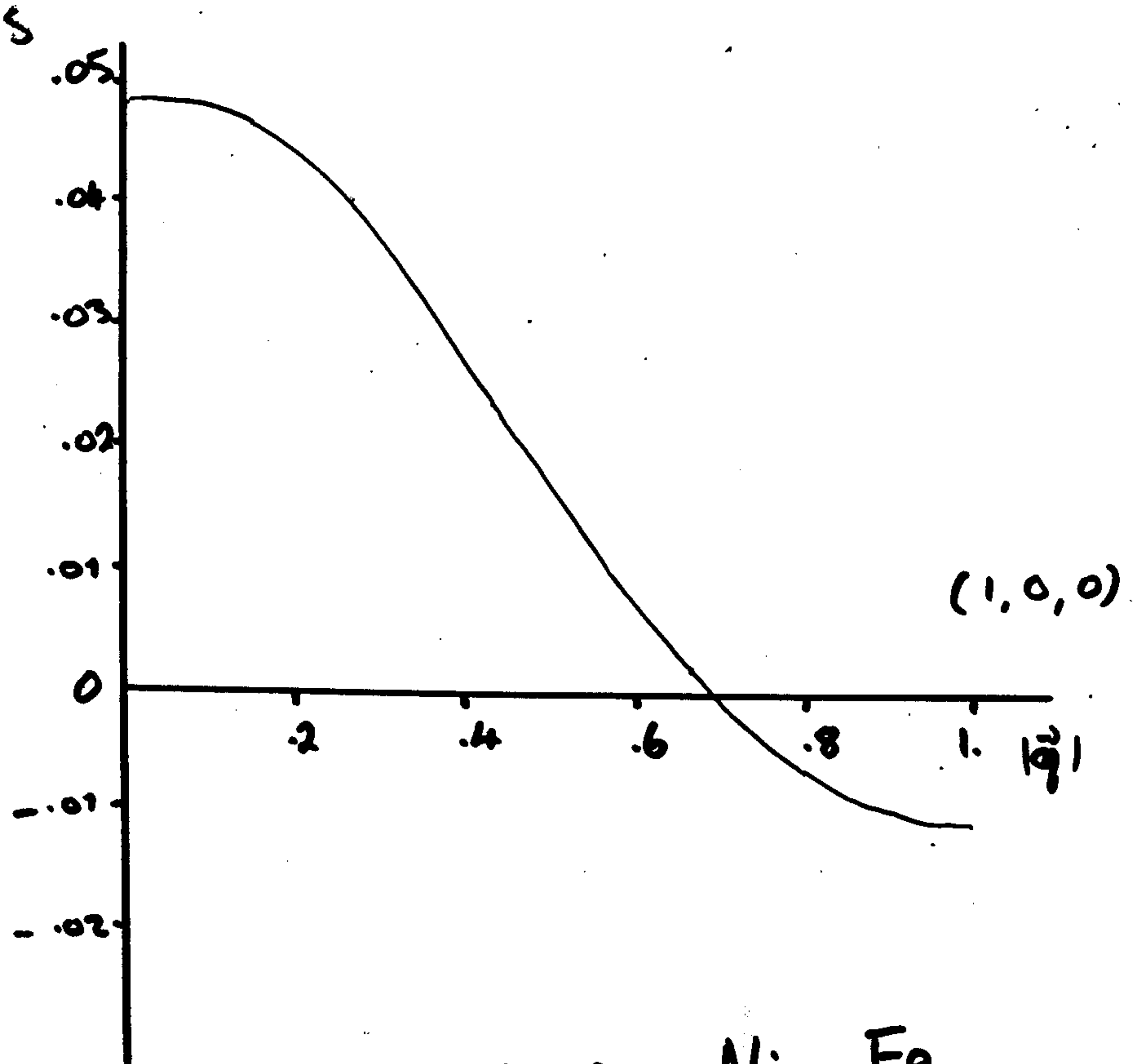
1. Compositional Correlations

$$\alpha(\vec{q}) = \beta c(1-c) \Big/ \left(1 - \beta c(1-c) S(\vec{q})\right)$$

$S(\vec{q})$ - dependent on electronic structure of paramagnetic state

2. Magnetic Correlations

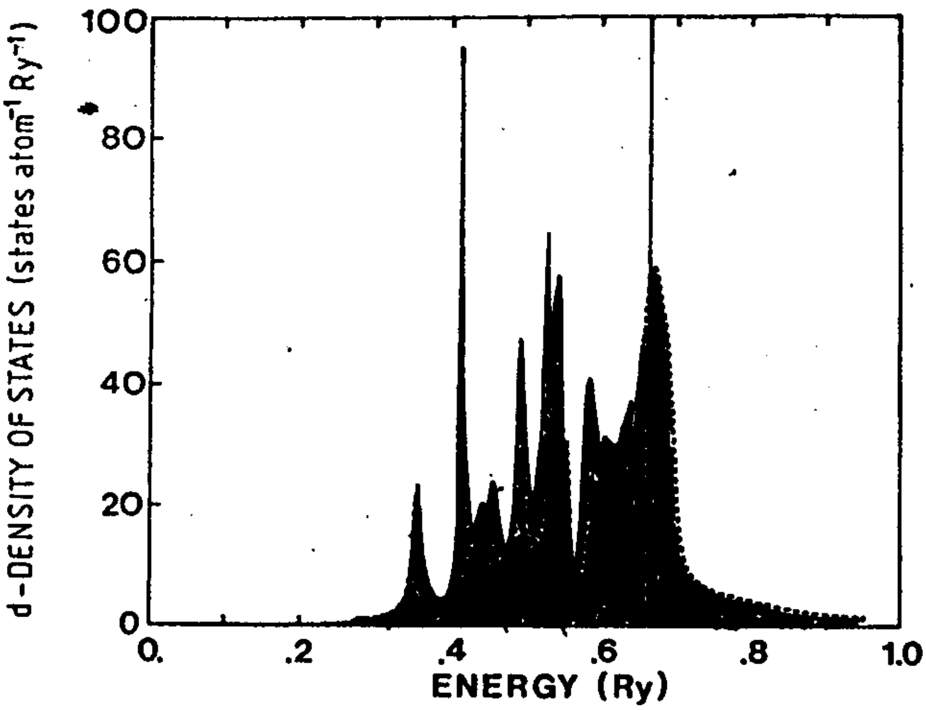
$$\chi(\vec{q}) = c\left(\chi_A^m(\vec{q}) + \chi_A^\mu(\vec{q})\right) + (1-c)\left(\chi_B^m(\vec{q}) + \chi_B^\mu(\vec{q})\right)$$



$S(\vec{q})$ $Ni_{.75}Fe_{.25}$
paramagnetic (Stoner)

Clustering

9.
12.



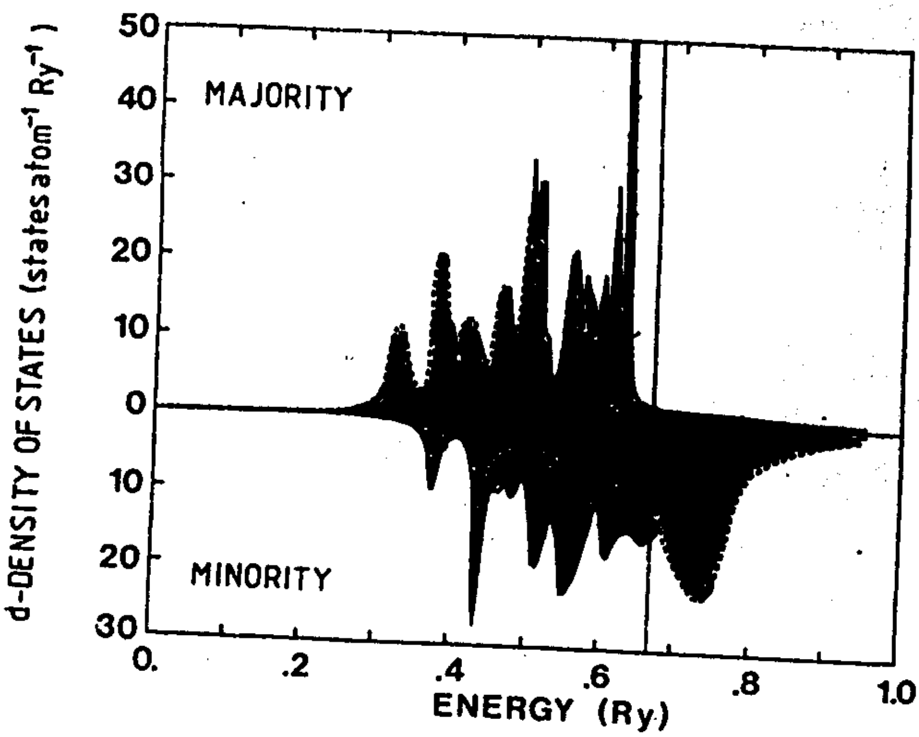
Paramagnetic

Ni_{75} Fe_{25}

Ni

Fe

Partially averaged d.o.s on Ni & Fe sites in Ni_{75} Fe_{25}



Ferromagnetic

Ni_{75} Fe_{25}

Ni

Fe

$Ni_{75}Fe_{25}$ (recall Stoner paramagnetic state)

Ordering correlations, v. low T_c in DLM
Interchange energy small
("local" exchange splitting on Fe sites)
$\bar{\mu}_{Fe} \approx 2.3\mu_B$, $\bar{\mu}_{Ni} \approx 0$

Ferromagnetic correlations found
$T_c^{mag.} > T_c^{comp.}$ .. system needs ferromagnetism to order

---

$Cu_{85}Mn_{15}$
Both Stoner and DLM paramagnetic states -- clustering correlations
Lower $T_c^{comp.}$ for DLM
(Some experimental evidence — ordering ??)
Antiferromagnetic correlations
$T_c^{mag.} \sim 150K$ into complicated magnetically ordered state
(agreement with experiment)

Summary

(i) General scheme:
Slowly varying fluctuations in itinerant electron systems
Onsager cavity fields

(ii) Paramagnetic state of ferromagnetic metals
Fe, Ni: reasonable
"first pass" theory

Range of behaviour in same framework.

(iii) Alloys: interrelation between magnetic and compositional ordering
<u>Cu</u>Mn, <u>Ni</u>Fe.