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NIST-DCS
Neutron Measurements of Lattice Disorder
in a Null-matrix ^{62}Ni-Pt Alloy Crystal and in a Dilute Ge-Si Crystal

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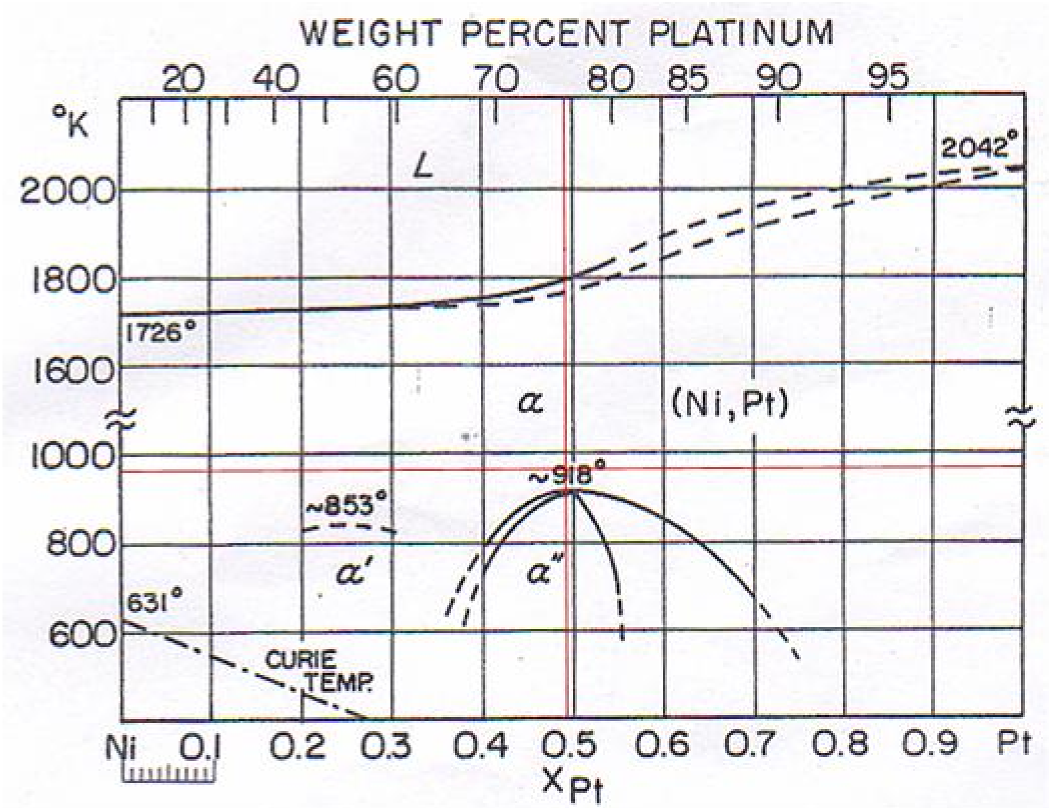
Null Matrix $^{62}Ni_{0.52}Pt_{0.48}$

NiPt Phase diagram



The NiPt sample was quenched Into water of 0°C from 700°C.

Same treatment for the Null matrix and Normal crystal

a_{Ni} = 3.52 Å
a_{Pt} = 3.92 Å

~ 11% difference

$$b_{^{62}Ni} = -8.7 \qquad b_{Pt} = 9.6$$

$$b_{Ni} = 10.3$$

$$c_{Ni} = 0.52 \qquad c_{Pt} = 0.48$$

for $\sin(\theta)/\lambda = 0$:

$$f_{Ni} = 27.99 \qquad f_{Pt} = 77.95$$

	$\lvert c_{Pt}b_{Pt}+c_{Ni}b_{Ni}\rvert^2$	$c_{Pt}c_{Ni}(b_{Pt}-b_{Ni})^2$
Null Matrix (Neutron)	0	83.59
Normal Crystal (Neutron)	99.28	0.12
X-ray (sin(θ)/λ=0)	$\lvert c_{Ni}f_{Ni}+c_{Pt}f_{Pt}\rvert^2$ = 2700	$c_{Pt}c_{Ni}(f_{Ni}-f_{Pt})^2$ = 623

Contrast Ratio: 696

Short-Range Order and Atomic Displacements

Q-space formalism (Krivoglaz)

$$I = I_{Bragg} + I_{Diffuse\,(D)} \quad : \text{standard}$$

$$I_D/N = c_A c_B (b_A - b_B)^2 \sum_{\mathbf{n}=0} \alpha_{\mathbf{n}} e^{i\mathbf{Q}\cdot\mathbf{r}_n}$$

$$= c_A c_B (b_A - b_B)^2 \alpha_{\mathbf{q}}$$

where $\mathbf{q} = \mathbf{Q} - \mathbf{G}_{hkl}$

α_n: pair correlations (Warren-Cowley SRO parameters)

$$c_A c_B \alpha_{\mathbf{q}} = N\langle|c_{\mathbf{q}}|^2\rangle$$

where $\langle|c_{\mathbf{q}}|^2\rangle$ is the Fourier transform of $\langle\sigma_i\sigma_j\rangle$,

and $\mathbf{r}_n \Rightarrow \mathbf{r}_n + \boldsymbol{\delta}_n$, then $\boldsymbol{\delta}_{\mathbf{q}} = \mathbf{A}_{\mathbf{q}} c_{\mathbf{q}}$, i.e. displacements dress local order

$$I_D/N\,(\mathbf{Q}) = \langle|c_{\mathbf{q}}|^2\rangle \times |\Delta b - \bar{b}\mathbf{Q}\cdot\mathbf{A}_{\mathbf{q}}|^2 \qquad \text{Total}$$

$$\langle|c_{\mathbf{q}}|^2\rangle \times |\Delta b|^2 \qquad \text{SRO}$$

$$\langle|c_{\mathbf{q}}|^2\rangle \times |\bar{b}\mathbf{Q}\cdot\mathbf{A}_{\mathbf{q}}|^2 \qquad \text{HDS}$$

$$-\langle|c_{\mathbf{q}}|^2\rangle \times \Delta b \times \bar{b}\mathbf{Q}\cdot\mathbf{A}_{\mathbf{q}} \qquad \text{SE}$$

Problems: 1) null matrix SE $\Rightarrow$ 0 (In a null-matrix alloy Δb=0)

2) no species-dependent displacements.

Real Space treatment (Borie-Sparks and Dietrich-Fenzl).

The total Diffuse intensity is given by:

$$I_{diff} = I - I_{Bragg} = I_{SRO} + I_{SE} + I_{HDS} + I_{TDS}$$

where:

$$I_{Laue} = c_A c_B \left| b_A - b_B \right|^2$$

$$I_{SRO} = I_{Laue} \sum_n \alpha_n e^{i\mathbf{Q}\cdot\mathbf{r}_n}$$

Is the intensity due to concentration fluctuations. α_n are the Warren-Cowley Parameters

$$\alpha_n = \frac{<\sigma_0^l \sigma_n^m> - c_i c_j}{c_i(\delta_{ij} - c_j)}$$

$\left\langle \sigma_0^A \sigma_n^B \right\rangle$ Is a concentration correlation function

$$\boxed{I_{sro} \Rightarrow \left\langle \sigma_0^l \sigma_n^m \right\rangle \quad \text{Dietrich - Fenzl}}$$

l, m are A or B

$$I_{SE} = I_{Laue} Q \cdot \sum_n \gamma_n e^{i\mathbf{Q}\cdot\mathbf{r}_n}$$

Is the distortion-induced "size-effect" scattering, where $\boldsymbol{r}_n = \boldsymbol{r}_n - \boldsymbol{r}_0$

$$\gamma = I^{-1}_{Laue} \sum_{ij} b_i b_j < \sigma_0^l \sigma_n^m >< \mu_n^{lm} >$$

γ_n are the linear displacement parameters

$$\boxed{I_{SE} \Rightarrow \frac{\langle \sigma_0^l \mu_n^m \rangle}{c_i} \quad \text{Dietrich- Fenzl}}$$

$< \mu_n^{lm} >$ is the average relative displacements between two atoms, separated by $\boldsymbol{r}_n - \boldsymbol{r}_0$

$$I_{HDS} = I_{Laue} \mathbf{Q} \otimes \mathbf{Q} \cdot \sum_n \varepsilon_n e^{i\mathbf{Q}\cdot\mathbf{r}_n}$$

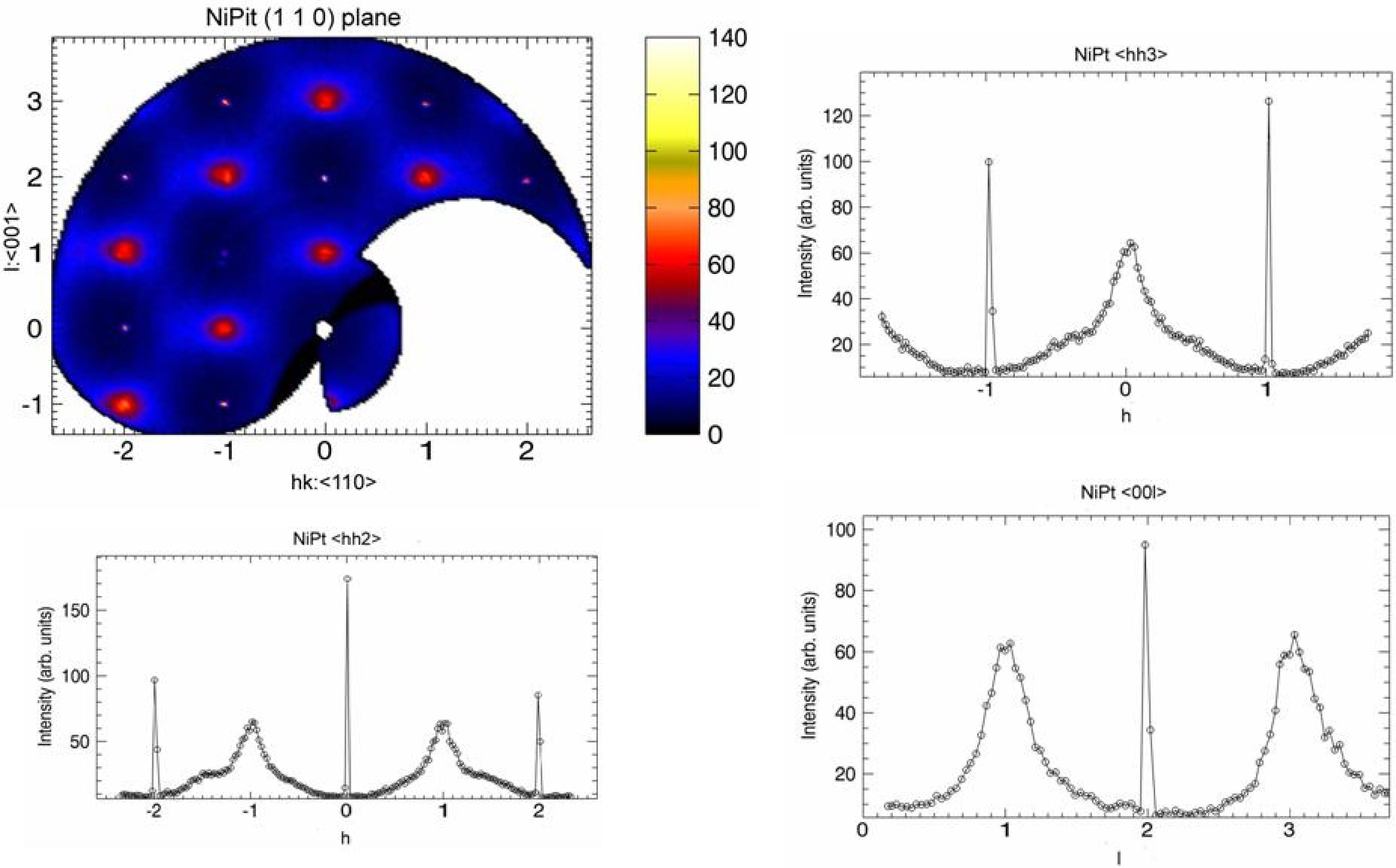
Is the quadratic term in both the displacements and scattering vector.

$$\varepsilon_n = I^{-1}{}_{Laue} \sum_{ij} b_i b_j^* \langle \sigma_0^l \sigma_n^m \rangle \langle (\mu \otimes \mu)_n^{lm} \rangle$$

$$\langle (\mu \otimes \mu)_n^{lm} \rangle = \frac{\langle \sigma_0^l \sigma_n^m (-\mu_0^l + \mu_n^m) \otimes (-\mu_0^l + \mu_n^m) \rangle}{\langle \sigma_0^l \sigma_n^m \rangle}$$

$$\boxed{I_{HDS} \Rightarrow \langle \mu_0^l \mu_n^m \rangle \quad \text{Dietrich - Fenzl}}$$

Null Matrix $^{62}Ni_{0.52}Pt_{0.48}$



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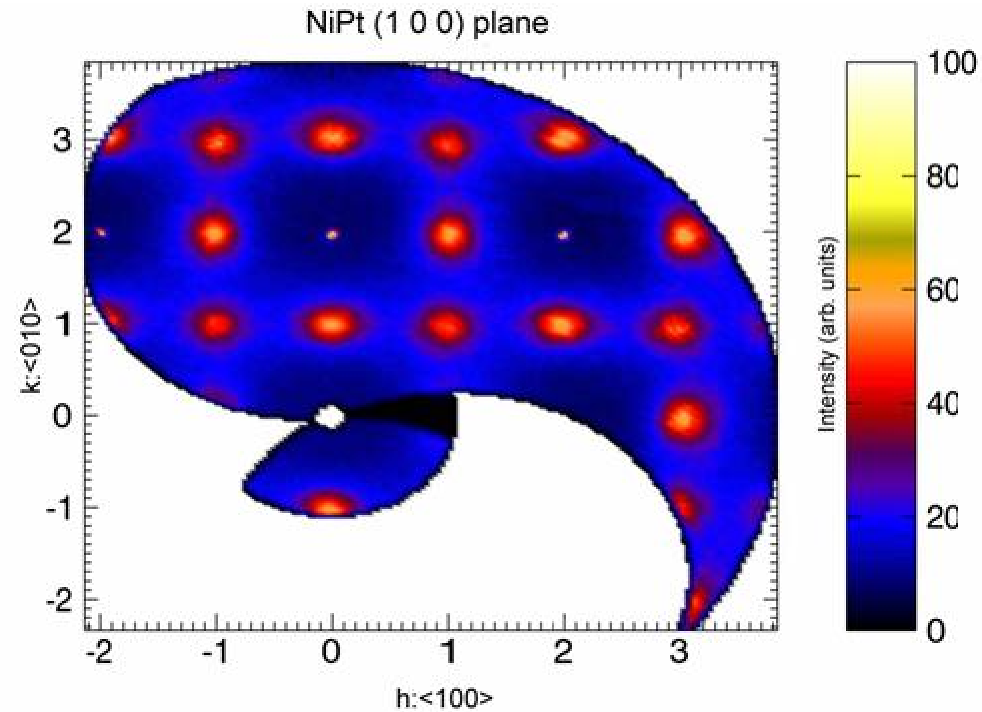

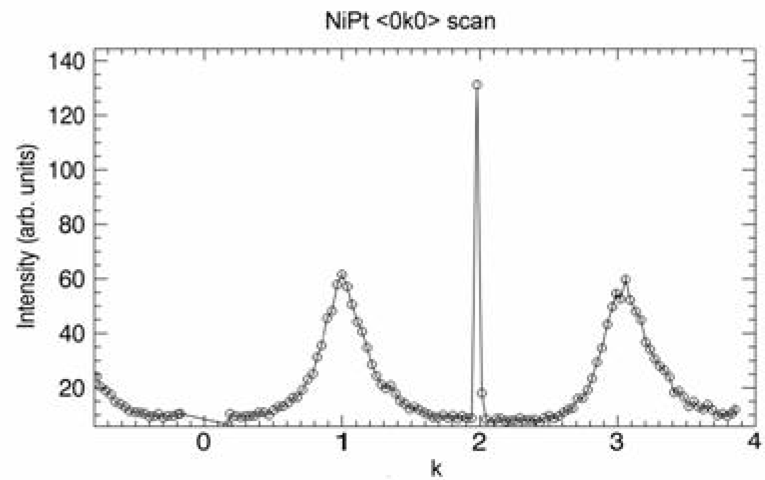

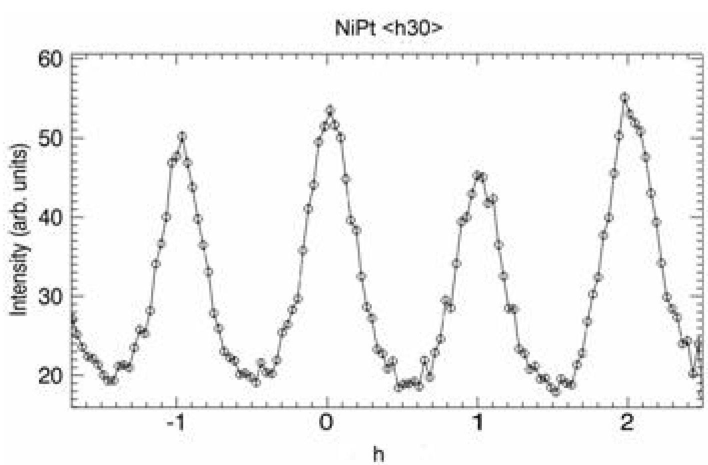

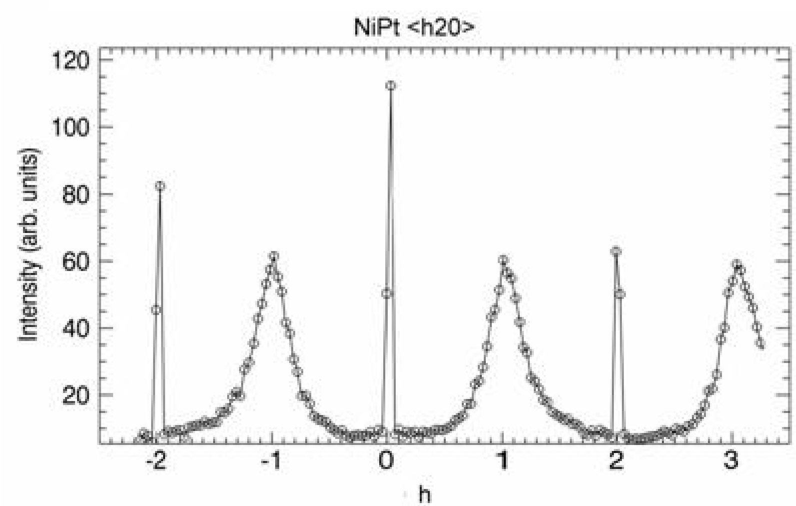

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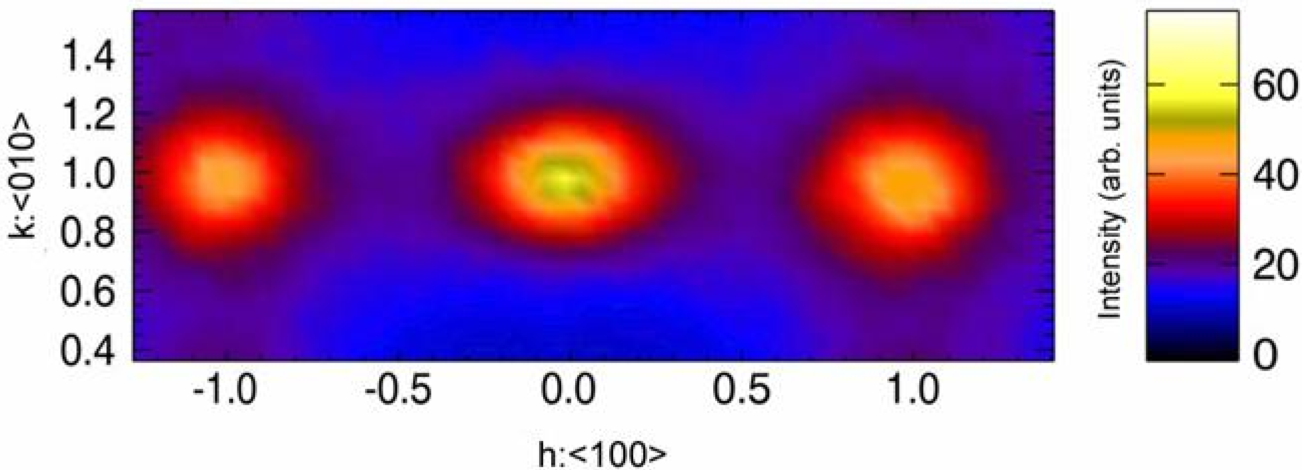
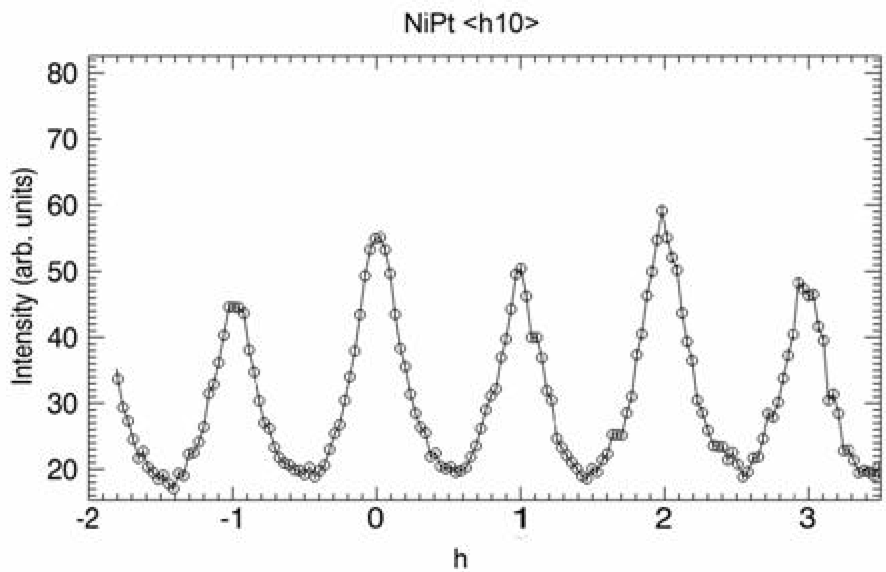
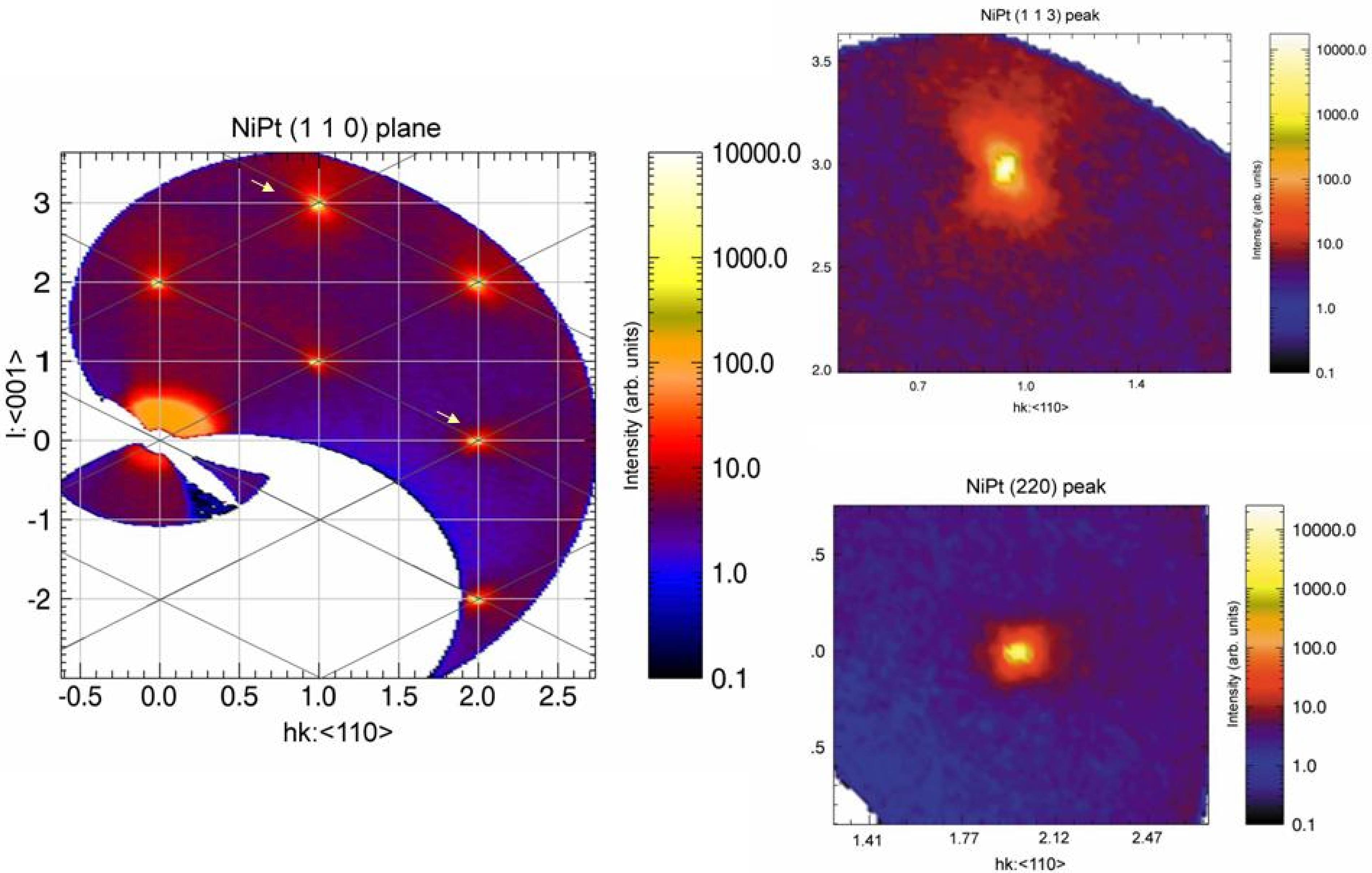




Normal Crystal $Ni_{0.52}Pt_{0.48}$



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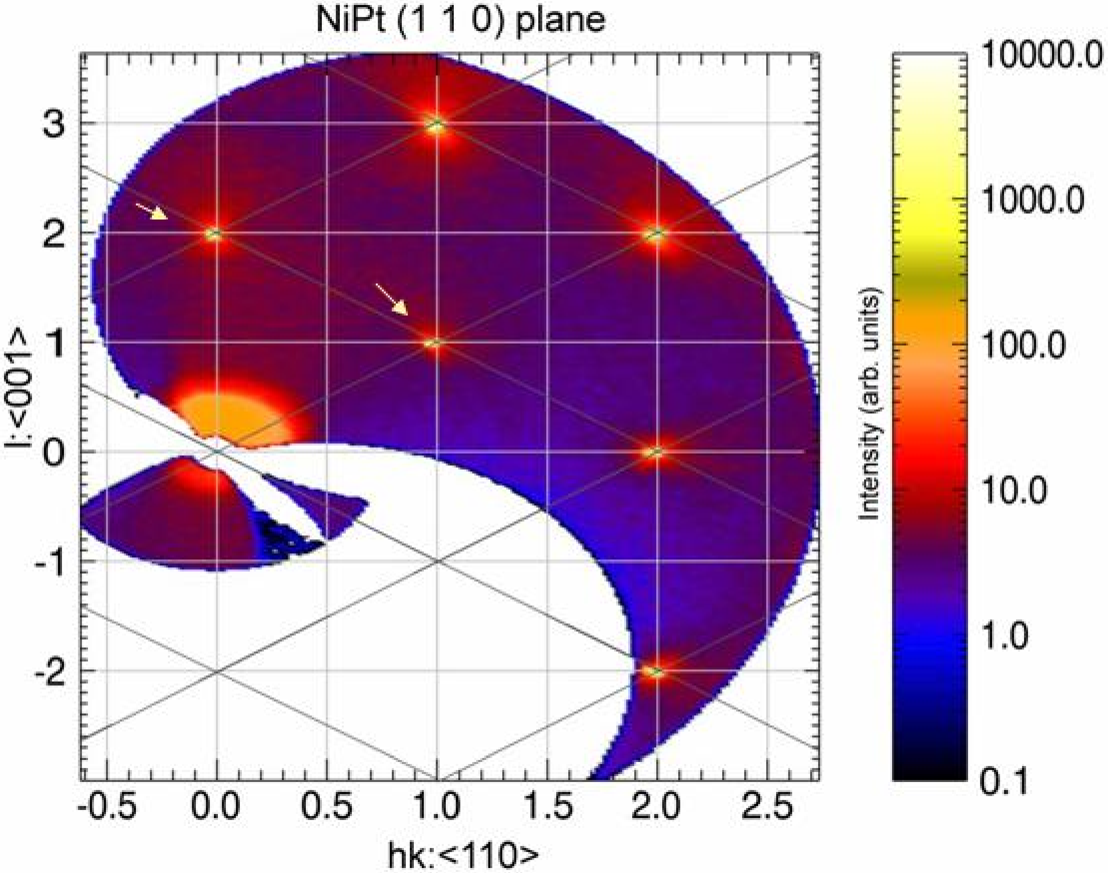
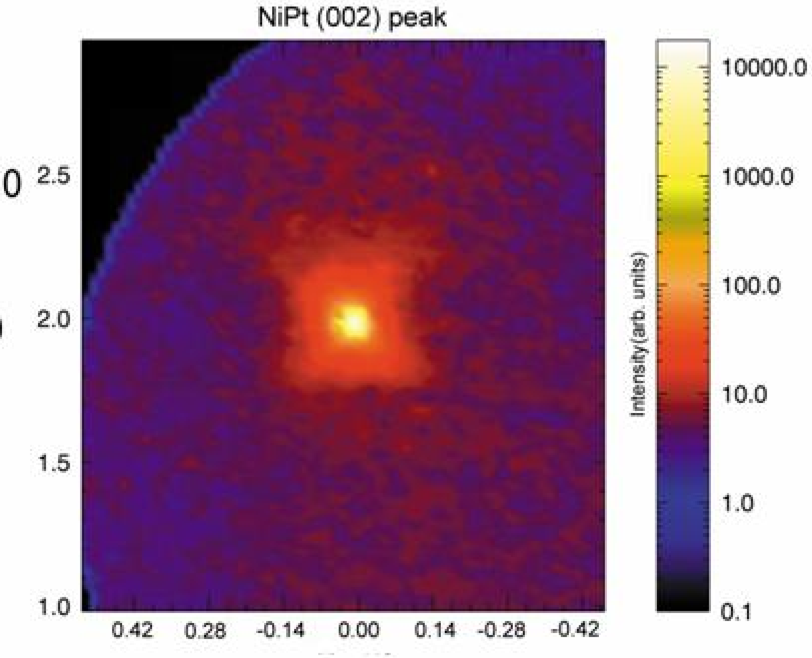
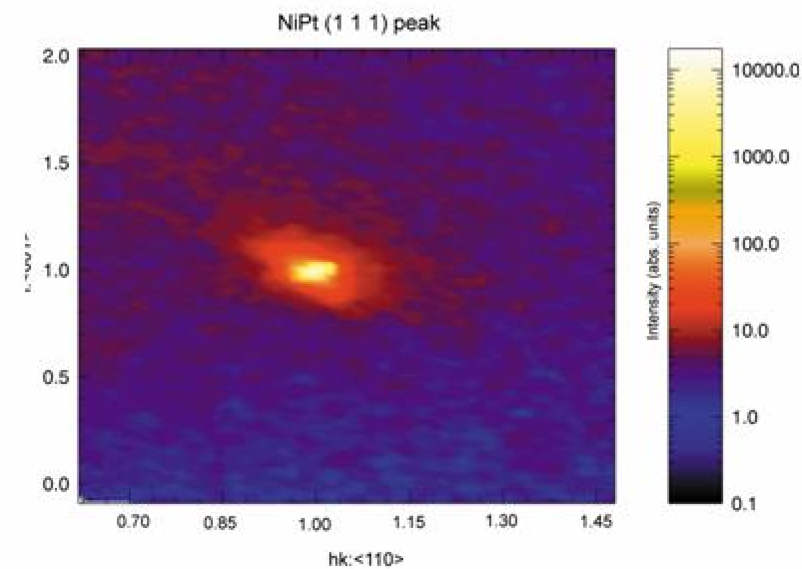


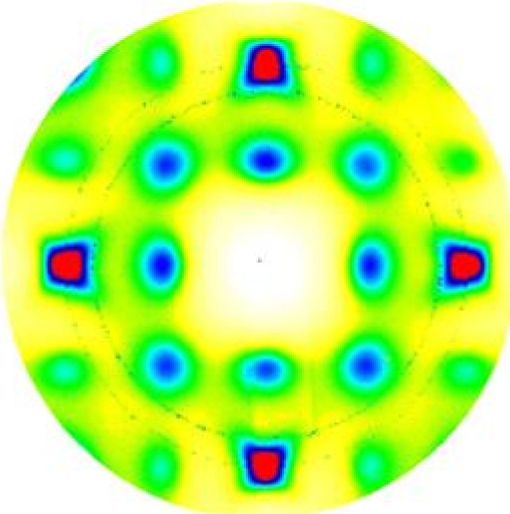




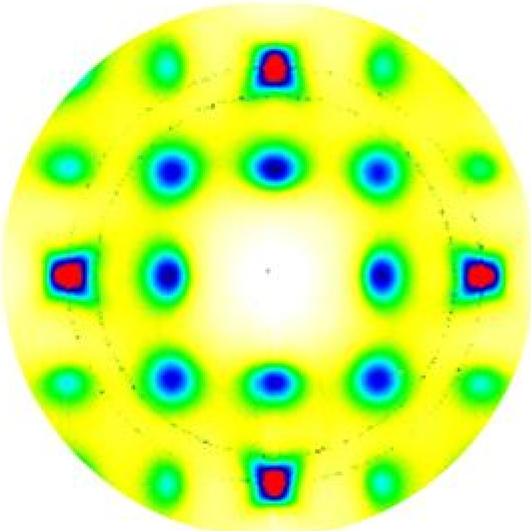
Normal Crystal $Ni_{0.52}Pt_{0.48}$

(X-ray Measurements)

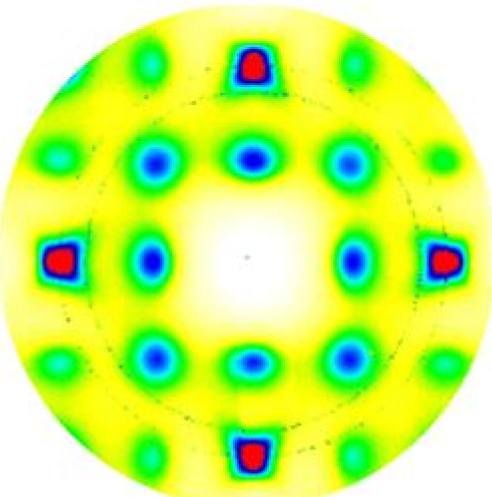
NiPt (100) Plane T = 800°C



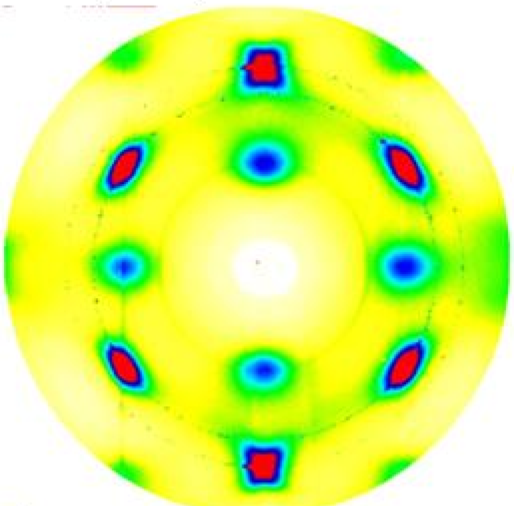
NiPt (100) Plane T = 720°C



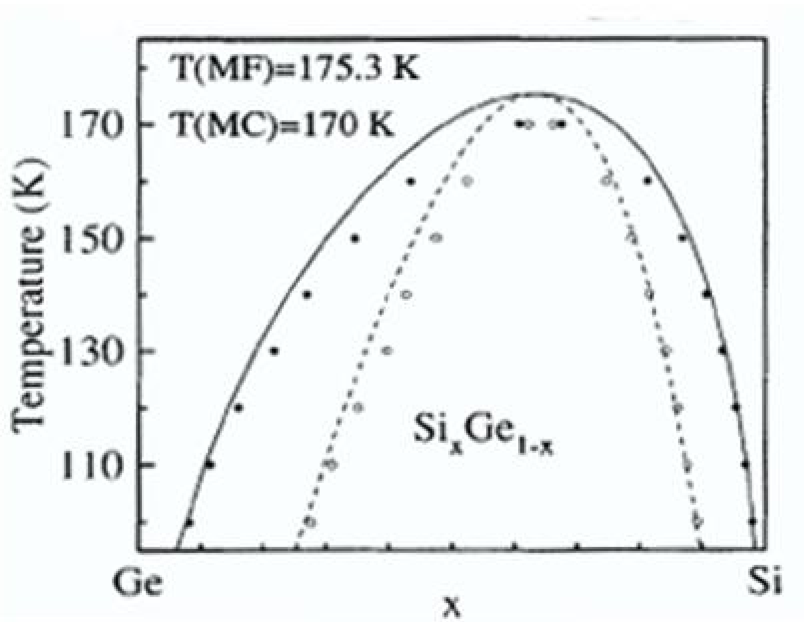
NiPt (100) Plane T = 750°C



NiPt (110) Plane T = 800°C

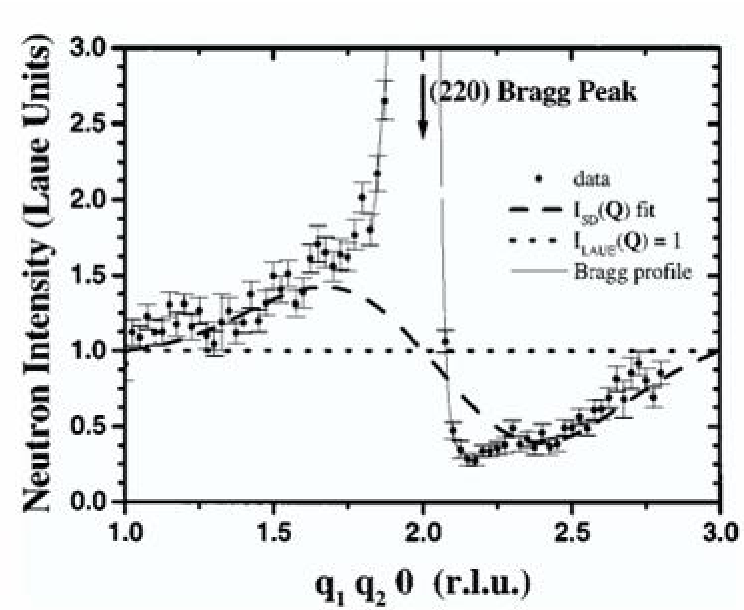


$Si_{0.92}Ge_{0.08}$ Single Crystal



$Si_x\ Ge_{1-x}$ Phase Diagram

S. De Gironcoli, P. Giannozozzi, and S. Baroni, Phys. Rev. Lett. 66, 2116 (1991)



Radial Scan through (220) using elastic neutron scattering.

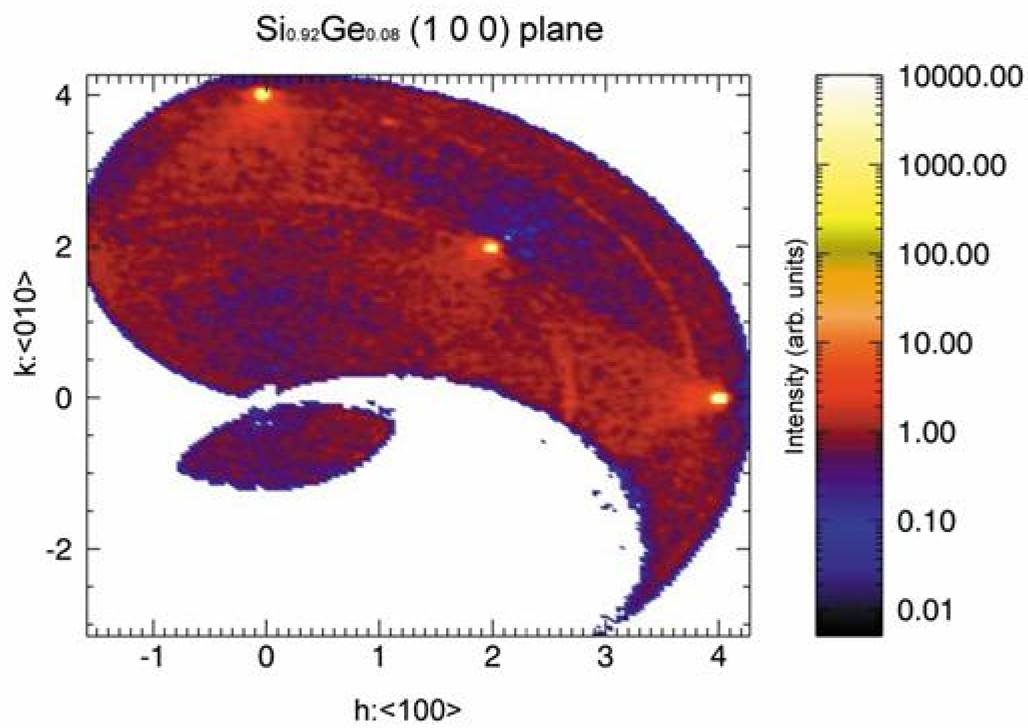
D. Le Bolloc'h, J. L. Robertson, H. Reichert, S.C. Moss and M. L. Crow, Phys. Rev. B 63 035204 (2001)

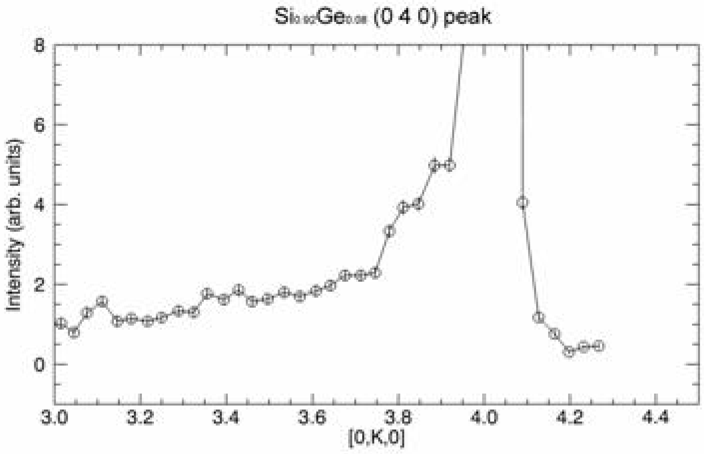
a_{Si} = 5.430 Å
a_{Ge} = 5.657 Å

~ 4% difference

$$b_{Si} = 4.1491$$
$$b_{Ge} = 8.185$$

$Si_{0.92}Ge_{0.08}$ Single Crystal





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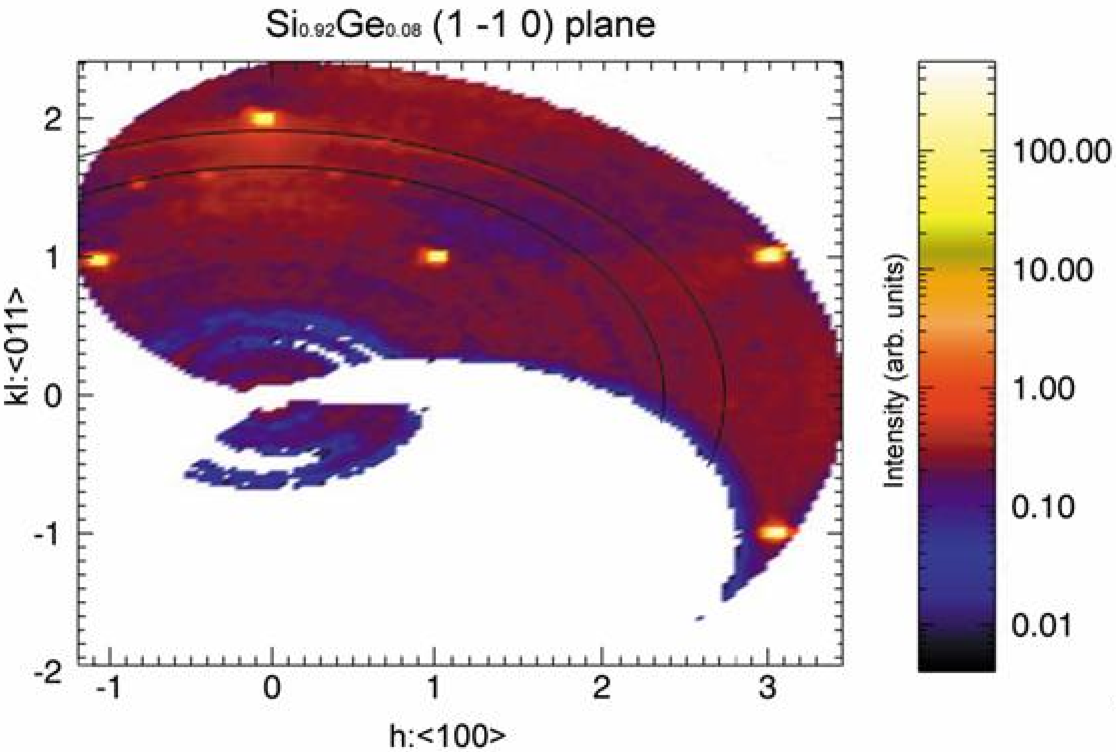

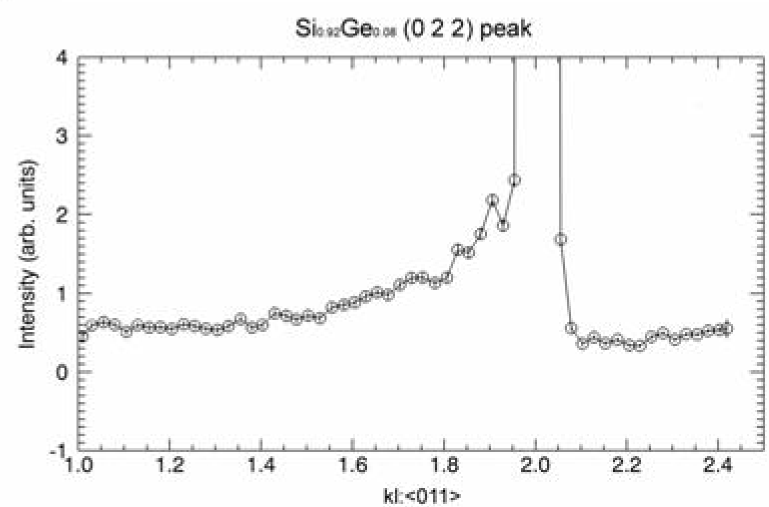

Why do we bother with this?

- SRO $\Rightarrow$ effective pairwise interactions – KCM Theory

  $<|c_{\mathbf{q}}|^2> = 1/(1+ 2c_A c_B \beta V_q)$ $\qquad \beta = 1/kT$

  $$V_{\mathbf{q}} = \sum_{\mathbf{m,n}} V_{\mathbf{m,n}} e^{\mathbf{iq \bullet r}_{mn}} \quad ; V_q = V_{\mathbf{q}}^{\mathbf{ch}} + V_{\mathbf{q}}^{\mathbf{si}}$$

  "chemical" "strain-induced"

  $V_{m,n} = \frac{1}{2}(V_{\mathbf{m,n}}^{AA} + V_{\mathbf{m,n}}^{BB} - 2V_{\mathbf{m,n}}^{AB})$; effective pairwise interactions

Theory gives us this or we supply theory

- SE + HDS $\Rightarrow$ sizes of atoms and their relative strain fields

- What is the "size" of an atom? Depends on "charge transfer" or "ionicity" and can take on many values in various alloys.

- The only experimental handle on phase stability in alloys is from the combination of electronic structure calculations and diffuse scattering of neutrons (or X-rays).