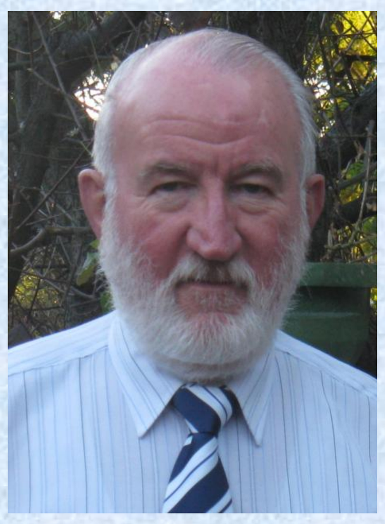
Diffuse Scattering Studies from an Fe-Pd Alloy

Trevor R. Finlayson[a], Garry J. McIntyre[b] and Kirrily C. Rule[b]

[a]Departments of Chemical Engineering & Physics, University of Melbourne, Victoria, 3010.
[b]Australian Centre for Neutron Scattering, ANSTO, Locked Bag 2001, Kirrowee, NSW, 2232.



Background

This project had its beginnings in the pioneering work of McIntyre *et al.* on the VIVALDI instrument at the Institute Laue-Langevin [1] to use the neutron Laue technique to map diffuse scattering and particularly low-energy, phonon scattering [2]. The availability of a large single crystal of Fe-Pd, of nominal composition Fe-30at%Pd, prompted our initial diffraction measurement on the KOALA instrument.

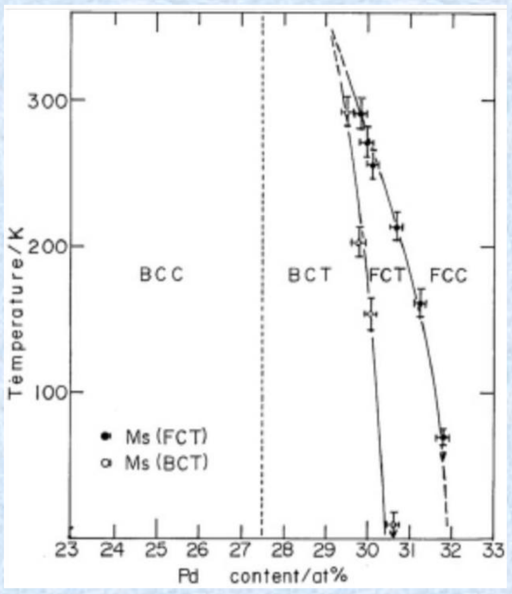


Figure 1. Part phase diagram for Fe-Pd system, showing the martensitic transformations. (After Sugiyama *et al.* [3].)

Relevant Properties of Fe-Pd

- For the composition range Fe-xat%Pd (where $30 \leq x \leq 32$), alloys transform martensitically, exhibiting quite complex structural behaviour on being cooled to low temperatures from room temperature (Figure 1).
- From x-ray diffraction studies, Seto *et al.* [4] suggested an "intermediate state" between the austenite (fcc) and martensite (fct) phases characterised by a "two-tetragonal-mixed" phase (Figure 2).
- From neutron scattering studies, the same authors [5] observed quasi-elastic scattering which increased as the crystal temperature approached the fcc to "intermediate phase" transition, (Figure 3) which they interpreted as "embryonic fluctuations" of the low-temperature structure.
- Earlier work [6] employing neutron inelastic scattering, suggested the fcc to fct transformation was driven by the softening of low-q, $[\zeta\zeta0][\zeta\bar{\zeta}0]$ (or TA_1) phonons (Figure 4).

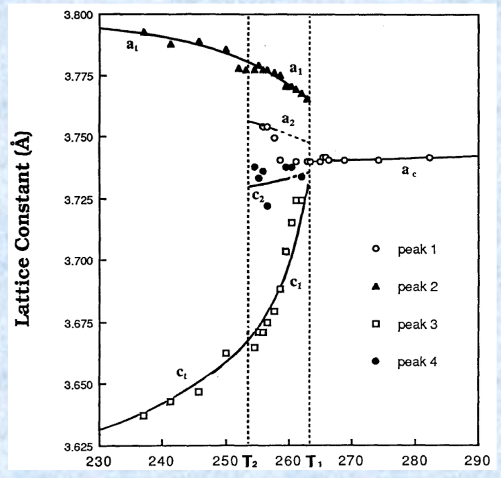


Figure 2. Temperature dependence of lattice constants calculated from peak positions around the (200) Bragg position in x-ray diffraction measurements, showing the "two-tetragonal-mixed" phase, suggested by Seto *et al.* [4] for an Fe-30.2at%Pd crystal.

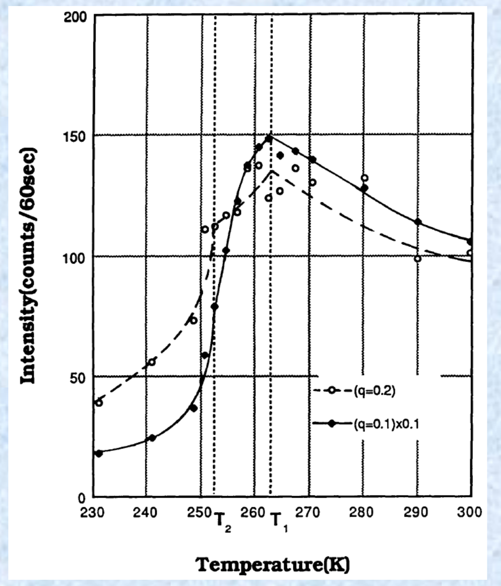


Figure 3. Temperature dependence of the intensity of the quasi-elastic scattering measured at (2.1, 1.9, 0) (full circles and solid line) and (2.2, 1.8, 0) (open circles and dashed line) for an Fe-30.2at%Pd crystal. (After Seto *et al.* [5].)

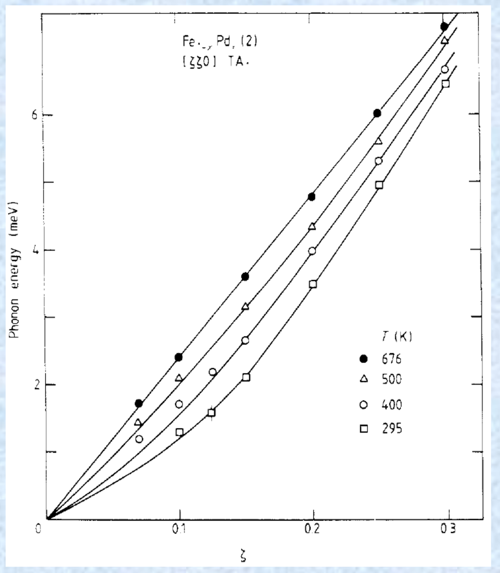


Figure 4. Temperature dependence of the $[\zeta\zeta0][\zeta\bar{\zeta}0]$ (or TA_1) phonon branch for an Fe-28at%Pd crystal which exhibited an fcc to fct transformation at 265K. (After Sato *et al.* [6].)

Aims of Current Project

(i) To study the diffuse scattering from the Fe-Pd crystal over the range $80\ K \leq T \leq 400\ K$;

(ii) To investigate the source of observed diffuse scattering.

Available Single Crystal

Nominally, Fe-30at%Pd in the form of an oblique cylinder, 7.7 mm high and diameter tapering from 10 mm to 12 mm (Figure 5). The parallel end faces are known to be {110}.

Crystal originally supplied by Dr. Ryuichiro Oshima, Osaka University, Japan, and machined from a large ingot grown by the Bridgman technique.

A triangular prism (~ 3 mm wide and 2 mm high) was electro-discharge machined from one edge of the crystal, as illustrated in Figure 5, making an ideal sample for a Laue experiment.

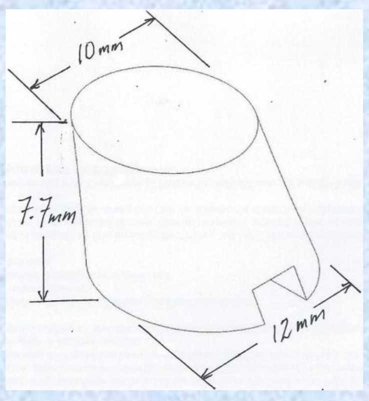


Figure 5. Sketch of Fe-30at%Pd single crystal. Used in the current project.

Preliminary Experiment

A Laue diffraction measurement was made at room temperature on the KOALA instrument, using the whole single crystal, with a 2 mm beam aperture. The result (Figure 6) showed a rich display of diffuse scattering, with the diffraction pattern being successfully indexed on the basis of the space group Fm3m with a_0 = 3.785 Å, d_{min} = 0.05 Å and λ_{min} = 0.77 Å.

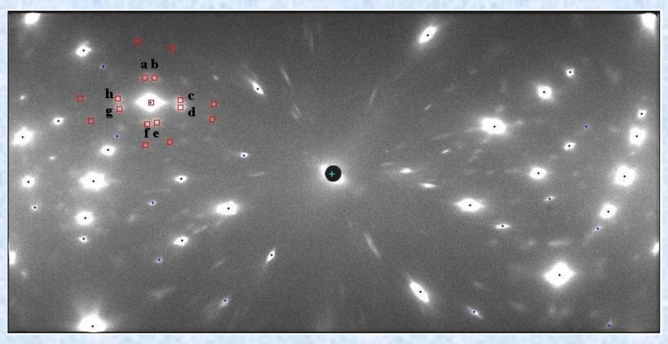


Figure. 6. Diffraction pattern from the whole crystal at room temperature, on the KOALA instrument. The major diffraction spot at about "10 o'clock" of the centre, is $(00\bar{l})$ (l = 2, 4, 6) with the $(00\bar{2})$ corresponding to λ = 2.47 Å. The eight labelled satellites have been indexed and averaging over the magnitudes of the two sets of indices, gives a likely propagation vector of 0.208 ±0.037 0 with $(00\bar{2})$ being the fundamental reflection, and the other satellites generated by a four-fold rotation. The outer spots with unlabelled small squares, are likely second-order satellites.

Current Experiments

Two experiments have been carried out at OPAL:

P6444 involved two days at KOALA (March, 2018) and eight days at TAIPAN (August, 2018); and

DB7608 involved one day at KOALA (May, 2019) and five days at SIKA (May, 2019).

Results: KOALA

While the transformations were evident from the temperature dependence of main diffraction spots from the ideally small crystal piece, (e.g., Figure 8), the interesting satellites highlighted in Figure 6 from the whole crystal were not observed, even after exposure times of 63 mins (Figure 7).

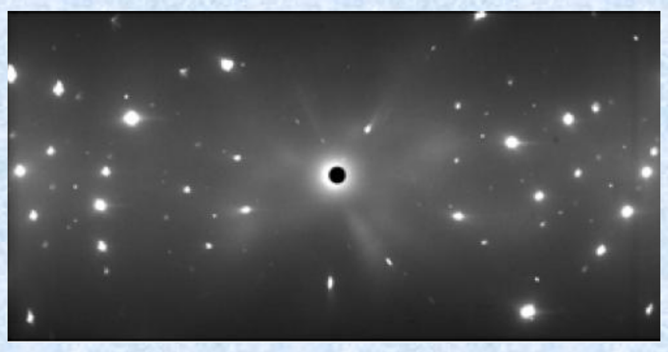


Figure 7. RT diffraction pattern from the small triangular prism piece of crystal.

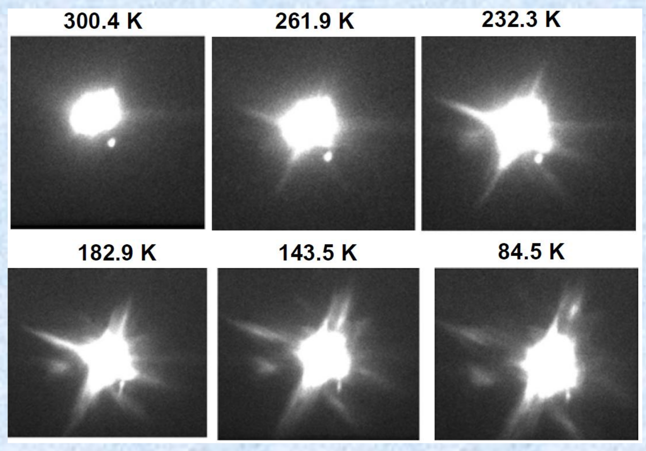


Figure 8. Temperature dependence of the $(\bar{2}00)$ diffraction spot at about "5 o'clock" of the centre in Figure 7.

Results: TAIPAN

Temperature dependences for elastic mesh scans around $(00\bar{2})$ and $(1\bar{1}1)$ reflections in hhl scattering plane. The transformations in the crystal were evident (e.g., Figure 9).

No evidence for interesting diffuse satellites.

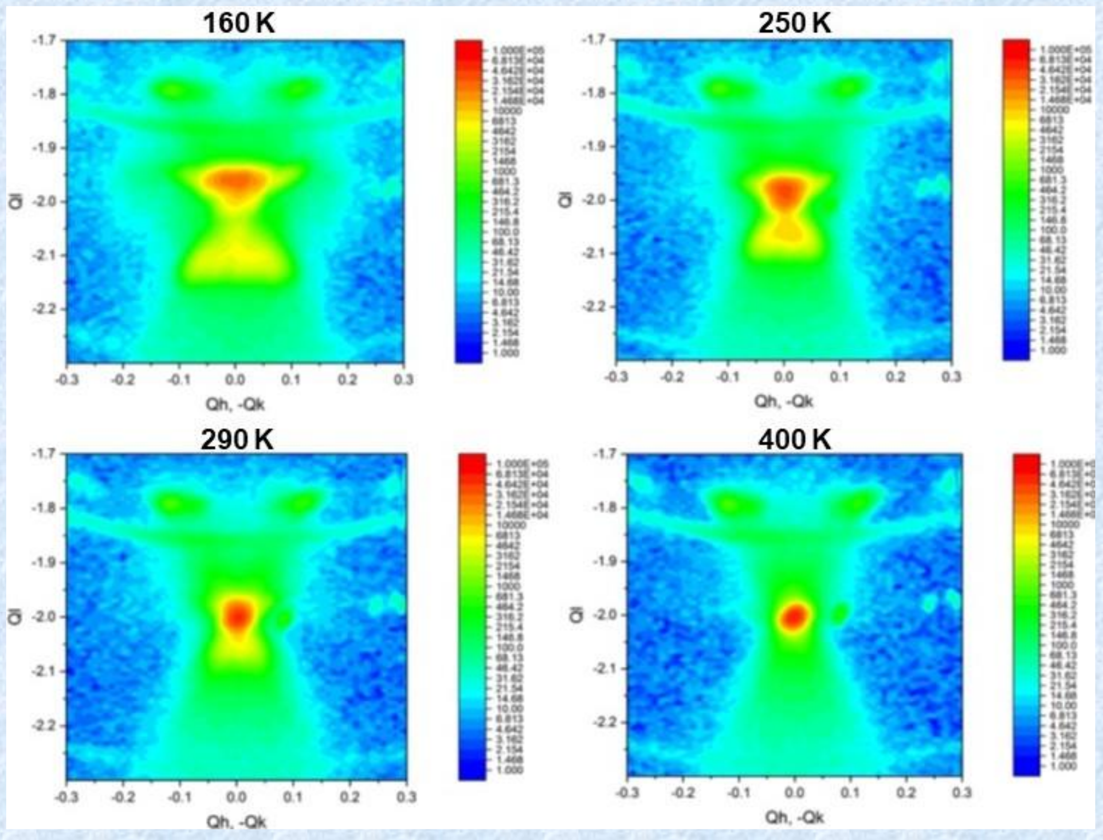


Figure 9. Elastic mesh scans around the $(00\bar{2})$ reflection.

Results: SIKA

Temperature dependence for elastic and inelastic mesh scans around (200) reflection in hk0 scattering plane and some TA_1 phonons.

First-order satellites observed and shown to involve elastic scattering to within 1 meV and the propagation vector around the (200) of -0.180 ±0.186 ±*0.034* (Figure 10).

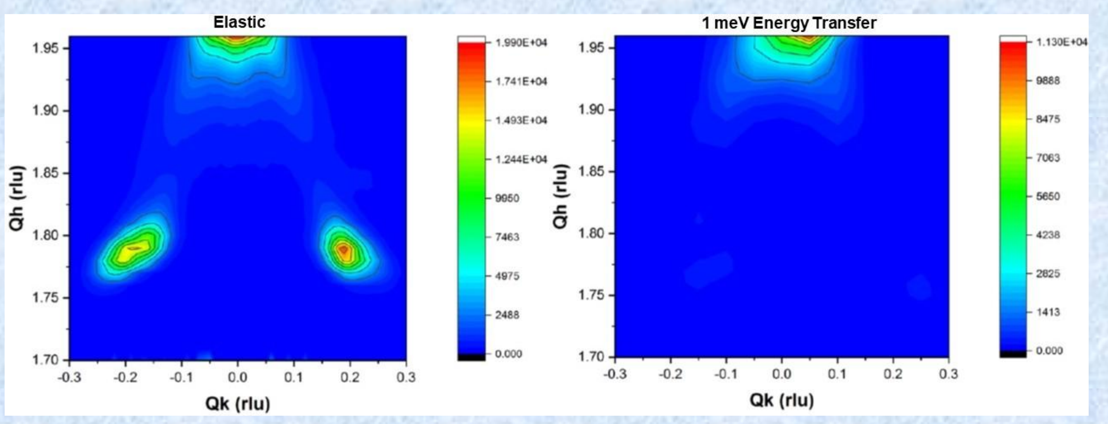


Figure 10. RT mesh scans around (200) reflection for elastic scattering (left) and 1 meV energy transfer (right), showing first-order satellites seen in initial KOALA experiment.

What Next?

What is the origin of these interesting satellites? Why have they not been observed from an ideally small piece of the crystal at KOALA? Hence, Proposal P9208 (September, 2020)?

References

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