Experimental report

05/01/2016

Proposal: 5-42-364 **Council:** 4/2014

Title: Small angle neutron scattering investigation of transverse magnetisation in INVAR Fe65Ni35

Research area: Physics

This proposal is a new proposal

Main proposer: John Ross STEWART

Experimental team: Sean GIBLIN
Jonathan TAYLOR
Toby WILLIS
John Ross STEWART

Local contacts: Dirk HONECKER

Samples: FeNi

Instrument	Requested days	Allocated days	From	To
D33	3	3	14/11/2014	17/11/2014

Abstract:

We propose to characterise ferromagnetic clusters in INVAR (Fe65Ni35) using polarised neutrons on D33. These clusters have been previously glimpsed in conventional and polarised neutron diffraction experiments, and dynamical evidence for them has been recently established by us, using muon spin relaxation and ferromagnetic neutron spin-echo. We intend to establish the longitudinal and transverse magnetisations associated with these FM clusters - and compare with non-collinear models of ferromagnetism in these alloys. We will also measure non-invar Fe60Ni40 to establish whether the nature of the FM clusters is integral to the invar effect. In all, we will need 3 days of beamtime on D33 to complete these measurements

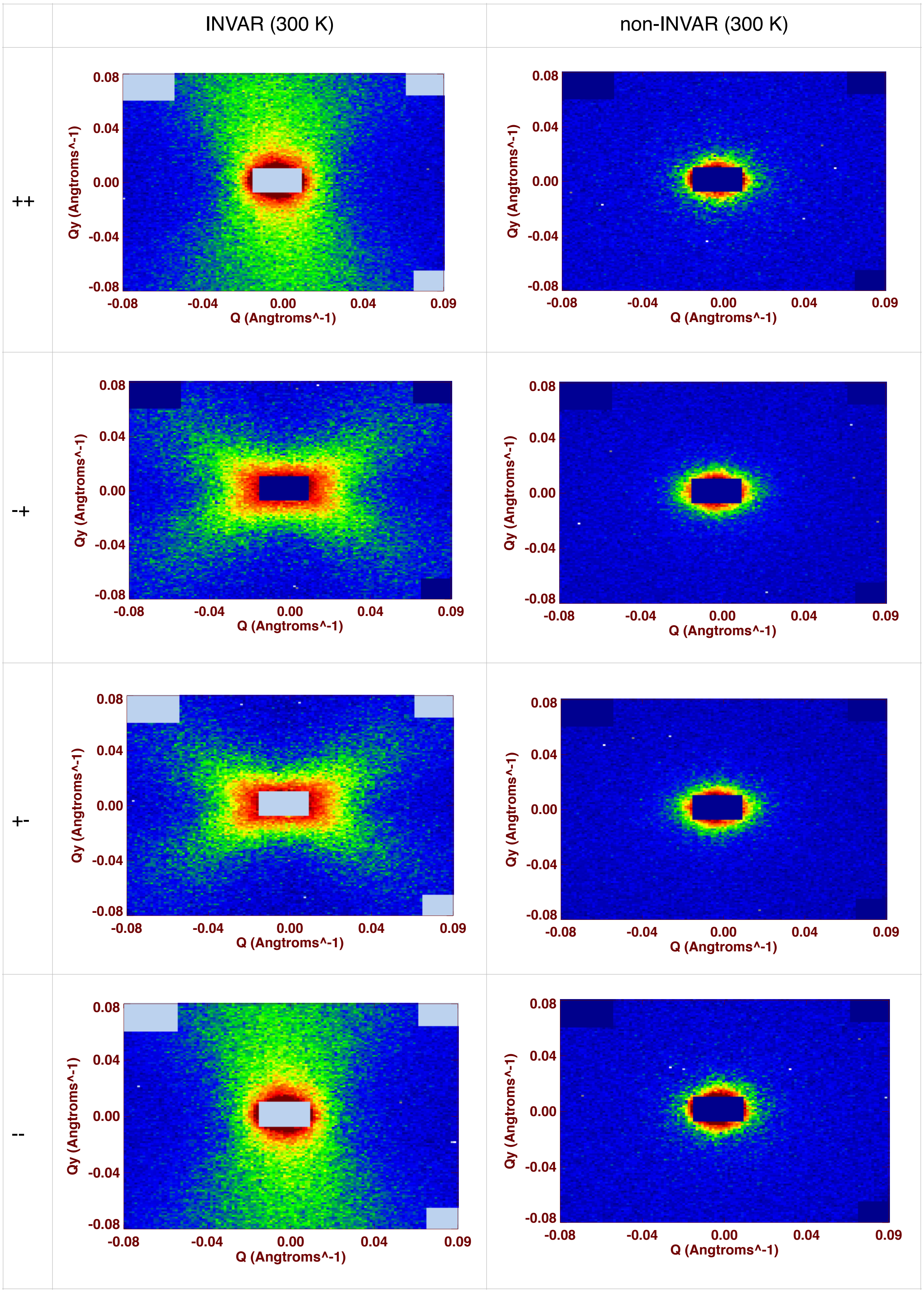
Since the "invar" effect was discovered in 1897 the mechanism producing negligible thermal expansion near room temperature has been widely debated. This effect is most commonly seen in ferromagnetic solid-solution alloys such as Fe-Ni and Fe-Pt, but the origin of the invar effect is still unknown. R J Weiss proposed a simple two state model[1], whereby thermal population of two nearly degenerate magnetic states exists in fcc Fe, a high spin (HS - 2.8 μ_B) high volume state and a low spin (LS - 0.5 μ_B) low volume state. The magneto-volume effect of this thermal population *possibly* acts to oppose the thermal expansion of the lattice. The electronic structure picture[2] of the HS to LS transition, whereby an electronic transfer between anti-bonding T_{2g} majority spin states and non-bonding E_g minority spin states results in the contraction of bonds, thereby counteracting the thermal expansion, provides a useful picture to understand the magneto-volume effect responsible for the near zero thermal expansion of the lattice in invar systems. However, at ambient pressure precise measurement of the ferromagnetic form factor using polarised neutron diffraction has shown that no such transfer occurs in Fe-Ni INVAR alloys[3].

There are a large number of separate ab-initio calculations of systems that exhibit the invar effect. Many of these make use of the Kohn–Korringa–Rostoker (KKR) greens function method, which is implicitly able to determine effects due to disorder in the system (Disordered Local Moment approach)[4]. Recent electronic structure calculations for disordered Fe-Pt[5] and $DyCo_2$ have shown a large negative volume magnetostriction, which presents a nice model for the observed thermal expansion anomalies. However results from other ab-initio calculations using similar methods compare less than favourably with experiment[6] in terms of both calculated and observed induced moment and overall spin momentum density distribution. There is a lack of experimental evidence that may or may not give credence to the role of disordered local moments in the INVAR phenomena.

Much has been also made of theoretical studies using state of the art ab-initio electronic structure calculations. Schilfgaarde[7] finds a non-collinear ferromagnetic ground state in INVAR concentrations of FeNi, and argues that the magnitude of the non-collinear moment couples to the atomic volume and explains the invar effect by virtue of this coupling having a negative (and counteracting) volume expansion on increasing temperature. Cowlam and Wildes[8] used neutron polarization analysis to show that INVAR $Fe_{65}Ni_{35}$ appears to be a simple collinear ferromagnet. However a preceding study by Menshikov and co-workers also using neutron polarisation analysis saw considerable diffuse scattering at small angles using the hot neutron polarised neutron diffractometer D5[9]. Crucially they noted that this "small angle scattering" appears only for the INVAR concentration, $Fe_{65}Ni_{35}$ and not "off-invar", and that the diffuse scattering was in the spin-flip channel indicating a considerable transverse magnetisation of around $\mu_\perp = 0.65\ \mu_B$ per atom. This could well be associated with non-collinear clusters - and perhaps lend credence to the Schilfgaarde model.

We have used the polarized neutron SANS diffractometer D33, in PA mode, to further characterise the magnetic clusters in a single crystal of INVAR, and one off-stoichiometry non-INVAR crystal, $Fe_{0.5}Ni_{0.5}$ in order to attempt to associate the INVAR effect with the presence and the characteristics of the magnetic clusters. The samples were cooled in a thin-tailed orange cryostat, and magnetic domains were aligned using a horizontal electromagnet at 0.58 T in the direction perpendicular to the beam. Data analysis, including corrections for finite polarization (spin-leakage) were performed using the LAMP package, and fitting done using IgorPro. The data taken for each spin-configuration were simultaneously fit to the standard "POLARIS" cross-sections as defined in the work of

Honecker and co-workers[1]. The spin-leakage corrections are in some doubt for this experiment as the initial polarization of the beam (taking account of the polarization transport through the transverse horizontal field) was not measured, but rather estimated from previous "ideal" characterisation of the initial polarisation. Figure 1 (below) shows the SANS data in each individual spin channel for the INVAR and non-INVAR crystals on the same normalised scale of intensity



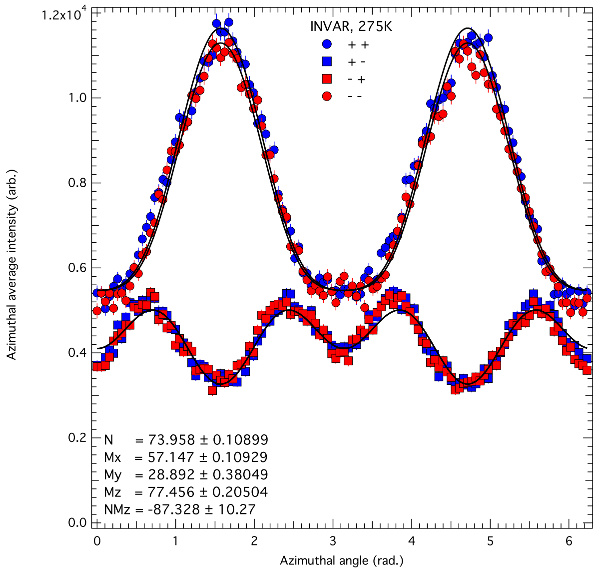
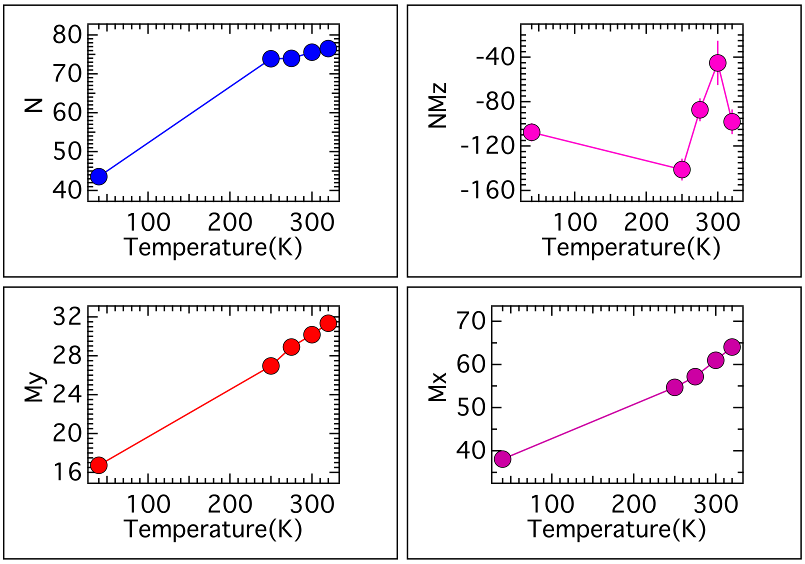


__Figure 2:__ Azimuthal average of INVAR spin-dependent SANS at 275 K. The lines are fits to the polarization dependent equations of Honecker[1].



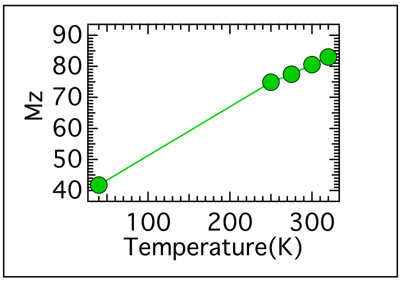


__Figure 3:__ Fit parameters for the INVAR sans data. N is the nuclear term, Mx, My and Mz are the square Fourier magnetisations in each direction, and NMz is the nuclear-magnetic interference term along the field direction (z)

The data clearly shows the presence of large static magnetic fluctuations in INVAR which are manifestly absent in non-INVAR. That these fluctuations are magnetic is clear from the azimuthal dependence of the SANS in Figure 1, and the spin-dependence of the data.

Fitting the azimuthally averaged data allows at least a partial separation of the nuclear and longitudinal (Mz) and transverse (My and Mz) square magnetisations in reciprocal space. Figure 3 shows the azimuthal dependence of the SANS in invar at 275 K fitted to the expressions of Honecker. The temperature dependence of the fitted parameters is shown in Figure 3. Some unresolved questions remain - particularly the question of why there is a temperature dependence in the nuclear part of the SANS. It is likely that high absorption of the rather thick single crystals is hampering an accurate subtraction of the background, making elucidation of the absolute nuclear scattering difficult. Notably, there also appears to be strong flat background in the spin-flip channels for both samples (which shows up in the Mx term for both invar and non-invar, and which shows no azimuthal dependence since the x-direction is along the beam).

While it is therefore likely that a continuation experiment will be need to check this, and too perform a crucial field scan to check the effect of imperfect domain alignment in the field, it is clear that the SANS in INVAR and non-INVAR are distinct, with clear signs of longitudinal and transverse static spin-fluctuations in INVAR.

---

[1] Honecker, D., Ferdinand, A., Döbrich, F., Dewhurst, C. D., Wiedenmann, A., Gómez-Polo, C., et al. (2010). Longitudinal polarization analysis in small-angle neutron scattering. The European Physical Journal B-Condensed Matter, 76(2), 209–213. http://doi.org/10.1140/epjb/e2010-00191-5