SPIN-LATTICE INTERACTIONS STUDIED BY POLARISED AND UNPOLARISED INELASTIC SCATTERING APPLICATION TO THE INVAR PROBLEM

P.J. BROWN
Institut Laue Langevin Avenue des Martyrs, 38042 Grenoble, France

ABSTRACT
A semi-quantitative analysis is given of some of the ways in which spin-lattice interactions can modify the cross-sections observable in neutron scattering experiments. This analysis is applied to the scattering from the invar alloy Fe₆₅Ni₃₅ using a model in which the magnetic moment is a function of the near neighbour separation. This model has been applied to clarify the results of inelastic scattering experiments carried out on Fe₆₅Ni₃₅ using both polarised and unpolarised neutrons. The extra information obtainable using polarised neutrons as well as the difficulties and limitations of the technique for inelastic scattering are discussed.

1. Introduction

Inelastic neutron scattering provides a powerful tool with which to study the interaction between the magnetic and vibrational degrees of freedom in crystals. When combined with a polarised incident beam and polarisation analysis of the scattered neutrons a maximum of information about the scattering process is obtained. Up to the present time the polarised technique has been little used mainly due to the difficulty of doing such experiments with relatively inefficient neutron polarisers. Recently however interest in the stability of magnetic moments in metals, and recognition of the important influence which the interatomic distances can have upon this has provided new motivation for such studies.

One of the better known examples of a macroscopic property mediated by spin-lattice interaction is the invar effect. In invar alloys the magnetic and lattice degrees of freedom interact in such a way that in a certain range of temperature, the invar temperature, the thermal expansion coefficient becomes very small. Although there is as yet no complete explanation of the effect it is generally agreed that around the invar temperature the increase in interatomic distance due to lattice vibrations is compensated by a decrease in the effective atomic volume brought about by a change in the magnetic state [1,2]. Recently it has become possible to carry out spin-polarised band structure calculations with reasonable accuracy. Two new calculations for the invar alloy Fe₆₅Ni₃₅ have been made [3,4] both of which indicate an incipient instability in the alloy at around the invar composition due to the existence of nearly degenerate states with different symmetries, magnetic moments and atomic volumes. It is supposed that it is the population and depopulation of these states driven by the lattice vibrations which gives rise to the invar property.
2. Cross-sections for inelastic magnetic scattering

The cross-sections for magnetic inelastic scattering of neutrons from an ordered magnetic material were first evaluated by Halpern and Johnson in their classical paper [5]. Here we use the form of the double differential cross-section given by Squires [6](p138) for spin-only scattering by moments localised at the nodes of a Bravais lattice:

\[
\frac{d^2\sigma}{d\Omega dE'} = (\gamma_n r_0)^2 \frac{k'}{k} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \kappa_{\alpha\kappa\beta}) \sum_{l} \sum_{l'} |F(Q)|^2 \delta(E - E' + \hbar\omega) \\
\times \sum_{\lambda\lambda'} p_{\lambda} \langle \lambda | S_\alpha^\beta \exp[-iQ \cdot R_l'] | \lambda' \rangle \langle \lambda' | \exp[iQ \cdot R_l] S_\beta^\beta | \lambda \rangle
\]  

(1)

Here \( \gamma_n \) is the neutron magnetic moment in nuclear magnetons and \( r_0 \) the classical electron radius; \( Q \) is the scattering vector, and \( \kappa \) a unit vector parallel to it. \( F(Q) \) is the magnetic form factor, \( R_l \) the position of the atom at lattice site \( l \) and \( S_\beta^\beta \) the \( \beta \) component of its spin. \( E \) and \( k \) are the energy and wavevector of the incident neutron, \( \lambda \) represents the initial state of the scattering system and \( p_{\lambda} \) its probability. The primed symbols are the final state values. The second line of Eq. 1 which contains the matrix elements for the scattering process can be written [6]

\[
\frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} \langle \hat{S}_\alpha^\beta(0) \exp(iQ \cdot \hat{R}_l(0)) \times \exp(-iQ \cdot \hat{R}_l(t)) \hat{S}_\beta^\beta(t) \rangle \exp(-i\omega t) dt
\]

(2)

where \( \hat{R}_l(t) \) and \( \hat{S}_\beta^\beta(t) \) are the time dependent Heisenberg operators

\[
\hat{R}_l(t) = \exp[i\hbar t/\hbar] R_l \exp[-i\hbar t/\hbar]
\]

(3)

\[
\hat{S}_\beta^\beta(t) = \exp[i\hbar t/\hbar] S_\beta^\beta \exp[-i\hbar t/\hbar]
\]

(4)

The angle brackets in Eq. 2 indicate the thermal average of the enclosed operator at the temperature of the system.

In order to see how the cross-sections are evaluated it is assumed to start with that there is no correlation of either the magnitudes or the directions of the magnetic moments of the atoms with their displacements \( u = \hat{R}_l - \hat{R}_l' \). In this case the thermal average of Eq. 1 can be factorised into the product of an average depending only on the atomic displacements and one depending only on magnetic fluctuations. Each of these averages is written as the sum of a constant part given by its value after infinite time, and a time-varying part. Thus

\[
\langle \exp[-iQ \cdot \hat{R}_l'] \exp[iQ \cdot \hat{R}_l(t)] \rangle = I_{l'l'}(Q, \infty) + I_{l'l'}(Q, t)
\]

(5)

and

\[
\langle \hat{S}_\alpha^\beta(0) \hat{S}_\beta^\beta(t) \rangle = J_{l'l'}^{\alpha\beta}(\infty) + J_{l'l'}^{\alpha\beta}(t)
\]

(6)

The product of Eq. 5 and Eq. 6 expands to give four terms:
(i) $I_{IV}(Q, \infty)J^{\alpha\beta}_{IV}(\infty)$ which is the cross-section for elastic magnetic scattering.

(ii) $I_{IV}(Q, t)J^{\alpha\beta}_{IV}(\infty)$ that for magneto-vibrational scattering. This term is elastic in the magnetic system and inelastic in the phonon system. For a ferromagnet with spins parallel to $z$ it gives scattering with the same $q$, $Q$ and $\omega$ dependence as phonon scattering by the atomic nuclei except that the $\sigma_{coh}/4\pi$ which occurs in the nuclear phonon cross-section is replaced by

$$\left(\frac{\gamma r_0}{2}\right)^2 |F(Q)|^2(1 - \kappa_z^2)(S^z)^2 = M_\perp^2$$ (7)

in the magneto-vibrational cross-section. Both cross-sections are proportional to

$$\frac{(\epsilon \cdot Q)^2}{\omega} \rightarrow \left[ \frac{(\epsilon \cdot Q)^2}{vq} \right]_{\text{lm}(Q \rightarrow 0)} \propto Q$$ (8)

where $\epsilon$ is the polarisation of the phonon, $v$ the phonon velocity and $q$ the reduced wavevector.

(iii) $I_{IV}(Q, \infty)J^{\alpha\beta}_{IV}(t)$ gives scattering inelastic in just the spin system. This can include excitations corresponding to both longitudinal fluctuations and transverse fluctuations (Spin waves).

(iv) $I_{IV}(Q, t)J^{\alpha\beta}_{IV}(t)$ scattering inelastic in both the phonon and spin systems.

3. Spin-lattice interaction in invar

If there is a significant spin-lattice interaction then the assumption that the magnetic state of an atom is independent of its displacement from the lattice node is no longer valid. There are several mechanisms which can give rise to such interactions; these include the Jahn Teller effect, quadropolar couplings, and magnetovolume effects. It is this latter mechanism which is probably important in invar alloys. All the recent electronic structure calculations for the Fe-Ni invar alloys, which are reviewed by [3], agree that there are states of nearly equal energy amongst which those with lower moment correspond to smaller atomic volumes. The different versions differ only in the actual composition of the states and on whether there are just two states or whether there is a continuum of states. For the present discussion these differences are unimportant and we shall adopt a very simple model in which we suppose that as a result of this peculiar band structure the atomic magnetic moment is a function of the nearest neighbour separation, or from the other point of view that there is a force between neighbouring atoms which depends on the magnitude of their moment. Suppose that this interaction leads to a linear dependence of the magnitude of the moment on the strain in the near neighbour bonds. Then as a first approximation the spin operators $\hat{S}$ can be renormalised so that

$$\hat{S}_i = \hat{S}_0 + \left(1 - \sum_{\alpha\beta} x_{\alpha\beta} e_{i_{\alpha\beta}}^\alpha \right)$$ (9)

where $x_{\alpha\beta}$ is a matrix of coefficients which depend on the physics of the magnetic moment fluctuation and $e_{i_{\alpha\beta}}^\alpha$ is the strain tensor at site $i$ due to the lattice vibrations.
It can be seen qualitatively that such a model will lead to additional terms in the inelastic scattering cross-section because it introduces a correlation between the moment fluctuations and the lattice displacements. If the electronic transitions which lead to the modification of the moment take place on a much shorter time-scale than the phonon frequencies the most important new contribution is to the magneto-vibrational scattering.

4. Magneto-vibrational scattering with spin-fluctuations driven by interatomic strain

Assuming the assumptions made above are valid and the renormalisation given by Eq. 9 is small the spin operators $S_i$ can be replaced by $S_{0l} \exp[-\xi_i]$ where $\xi_i$ is an operator which gives the fractional deviation in the magnitude of the moment at lattice site $l$ from its mean value. The dependence of the moment on the atomic displacements is now completely contained in the operator $\xi_i$ so it is again possible to factorise Eq. 5 but as the more complex expression

$$\left\langle \exp \left[ -iQ \cdot \hat{R}_{l}(0) - \hat{\xi}_l(0) \right] \exp \left[ iQ \cdot \hat{R}_l(t) - \hat{\xi}_l(t) \right] \right\rangle \times \langle S_0^{\beta} S_{0l}^{\beta} \rangle$$

and this may be developed to give four terms similar to those enumerated in section 3. The thermal average which determines the magneto-vibrational scattering becomes

$$\left\langle \exp \left[ -iQ \cdot \hat{R}_{l}(0) - \hat{\xi}_l(0) \right] \exp \left[ iQ \cdot \hat{R}_l(t) - \hat{\xi}_l(t) \right] \right\rangle \times \langle S_{0l}^{\beta} \rangle \langle S_{0l}^{\beta} \rangle$$

For a ferromagnet with spins parallel to $z$ the spin dependent term in the cross-section reduces to

$$\left( \frac{g n r_0}{2} \right)^2 \left| F(Q) \right|^2 \left( \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \kappa_\alpha \kappa_\beta) \langle S_{0l}^{\beta} \rangle \right)^2$$

$$= \left( \frac{g n r_0}{2} \right)^2 \left| F(Q) \right|^2 (1 - \kappa_z^2) \langle S_0^{\beta} \rangle^2 = M_0^2$$

The operators $\hat{R}_l = \hat{l} + \hat{u}_l$ can be expressed in terms of the phonon anihilation and creation operators $\hat{a}$ and $\hat{a}^+$ since

$$\hat{u}_l = \left( \frac{\hbar}{2m_\alpha N} \right)^{1/2} \sum_s \frac{\epsilon_s}{\omega_s} (\hat{a}_s \exp[iq_s \cdot \hat{l}] + \hat{a}_s^+ \exp[-iq_s \cdot \hat{l}])$$

where $m_\alpha$ is the mass of the atom and the sum is over the $N$ phonon modes with wave-vector $q_s$, frequency $\omega_s$, and polarisation $\epsilon_s$ in the 1st Brillouin zone. The strain operator $\partial \hat{u}/\partial \hat{l}$ can also be written in terms of the operators $\hat{a}$ and $\hat{a}^+$

$$\hat{\xi}_l = \left( \frac{\hbar}{2m_\alpha N} \right)^{1/2} \sum_s \sum_{\alpha,\beta} \frac{x_{\alpha,\beta} q_s^2 \epsilon_s^\beta}{\sqrt{\omega_s}} \left[ (\hat{a}_s \exp[iq_s \cdot \hat{l}] - \hat{a}_s^+ \exp[-iq_s \cdot \hat{l}] \right]$$

and we define

$$\Xi_s = \sum_{\alpha,\beta} x_{\alpha,\beta} q_s^2 \epsilon_s^\beta$$
Following [6] (p29) we write the thermal average

\[
\langle \exp \left[ -i\mathbf{Q} \cdot \hat{R}_l(0) - \hat{\xi}_l(0) \right] \exp \left[ i\mathbf{Q} \cdot \hat{R}_l(t) - \hat{\xi}_l(t) \right] \rangle = \exp \left[ -i\mathbf{Q} \cdot (l' - l) \right]\langle \exp U \exp V \rangle
\]

with

\[
U = -i\mathbf{Q} \cdot \hat{R}_l(0) - \hat{\xi}_l(0) = -\sum_s \hat{a}_s(g_s + \gamma_s) + \hat{a}_s^+(g_s - \gamma_s)
\]

\[
V = i\mathbf{Q} \cdot \hat{R}_l(t) - \hat{\xi}_l(t) = \sum_s \hat{a}_s(h_s - \eta_s) + \hat{a}_s^+(h_s^* + \eta_s^*)
\]

\[
g_s = \left( \frac{\hbar}{2m_aN} \right)^{1/2} \frac{Q \cdot \epsilon_s}{\sqrt{\omega_s}}, \quad \gamma_s = \left( \frac{\hbar}{2m_aN} \right)^{1/2} \frac{\Xi_s}{\sqrt{\omega_s}}
\]

\[
h_s = \left( \frac{\hbar}{2m_aN} \right)^{1/2} \frac{Q \cdot \epsilon_s}{\sqrt{\omega_s}} \exp(i(q \cdot l - \omega_st))
\]

\[
\eta_s = \left( \frac{\hbar}{2m_aN} \right)^{1/2} \frac{\Xi_s}{\sqrt{\omega_s}} \exp(i(q \cdot l - \omega_st))
\]

The magneto-vibrational cross-section can then be expanded

\[
\frac{d^2\sigma}{d\Omega dE'}_{MV} = \frac{k'}{k} \frac{N}{2\pi\hbar} M_{\perp}^2 \exp \langle U^2 \rangle \sum_l \exp[i\mathbf{Q} \cdot \mathbf{l}]
\]

\[
\times \int_{-\infty}^{\infty} \exp \langle UV \rangle \exp[-i\omega t] dt
\]

\[
\langle U^2 \rangle = -\sum_\lambda \sum_{s,s'} \langle \lambda | (\hat{a}_s(g_s + \gamma_s) + \hat{a}_s^+(g_s - \gamma_s))
\]

\[
\times (\hat{a}_{s'}(g_{s'} + \gamma_{s'}) + \hat{a}_{s'}^+(g_{s'} - \gamma_{s'})) |\lambda\rangle
\]

\[
= -\sum_\lambda \sum_s (g_s^2 - \gamma_s^2) \langle \lambda | \hat{a}_s \hat{a}_s^+ + \hat{a}_s^+ \hat{a}_s |\lambda\rangle
\]

so that the Debye Waller term becomes

\[
\exp \langle U^2 \rangle = \exp \left[ -\frac{\hbar}{2m_aN} \sum_s \frac{(Q \cdot \epsilon_s)^2 - \Xi_s^2}{\omega_s} (2n_s + 1) \right] = \exp(-2W')
\]

where \(\langle n_s \rangle\) is the probability of the state \(n_s\) at the temperature of the system. The one-phonon cross-section is obtained from the term in \(\langle UV \rangle\) in the expansion of \(\exp(UV)\).

\[
\langle \lambda | UV | \lambda \rangle = \sum_{s,s'} \langle \lambda | (\hat{a}_s(g_s + \gamma_s) + \hat{a}_s^+(g_s - \gamma_s))
\]

\[
\times (\hat{a}_{s'}(h_{s'} - \eta_{s'}) + \hat{a}_{s'}^+(h_{s'}^* + \eta_{s'}^*)) |\lambda\rangle
\]
giving

\[
\langle U\dot{V}\rangle = \sum_s (g_s + \gamma_s) (\hbar^+_s + \eta^+_s) (n_s + 1) + (g_s - \gamma_s) (\hbar_s - \eta_s) (n_s)
\]

\[
= \left(\frac{\hbar}{2m_a N}\right) \sum_s \frac{1}{\omega_s} \left((Q \cdot \epsilon_s + \Xi_s)^2 \exp[-i(q \cdot l - \omega_st)] (n_s + 1) \right.
\]

\[
+ (Q \cdot \epsilon_s - \Xi_s)^2 \exp[i(q \cdot l - \omega_st)] (n_s) \right)
\]  

(24)

The cross-section for magneto-vibrational scattering by one phonon creation is thus

\[
\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{M^+} = \frac{k'}{k} \frac{(2\pi)^3}{2m_a v_0} \frac{M^2_{Q \perp}}{\omega_s} \sum_s \sum_{\tau} \frac{(Q \cdot \epsilon_s + \Xi_s)^2}{\omega_s} (n_s + 1)
\]

\[
\times \delta(E - E' - \hbar\omega_s) \delta(Q + q - \tau)
\]  

(25)

where \(\tau\) is a reciprocal lattice vector.

5. Inelastic experiments with polarised neutrons

5.1 The polarised neutron cross-sections

The full range of cross-sections which can be measured on a triple-axis spectrometer with a polarising monochromator and analyser were discussed by [7]. Rather more information can be obtained if it is possible to measure all three components of the scattered polarisation rather than just the \(z\)-component [8]. General expressions for the cross-section for polarised neutrons and for the polarisation of the scattered beam are given in [9]. In the experiments to be described here an unpolarised incident beam was scattered from a magnetised sample and the separate intensities of neutrons scattered with spins parallel and antiparallel to the magnetisation direction were measured. So as not to make the expressions too complicated the discussion will be limited to this special case. The double differential cross-section for joint nuclear and magnetic scattering has the form

\[
\frac{d^2\sigma}{d\Omega dE'} \propto \sum_{l'} \sum_{\sigma\sigma'} p_{l'} p_{\lambda'} \left| \left\langle \lambda\sigma \mid \exp[-iQ \cdot R_{l'}](\hat{T}_{l'} \cdot \hat{\sigma} + \hat{M}_{Q l'} \cdot \hat{\sigma}) \right| \lambda'\sigma' \right|^2
\]  

(26)

Here \(\hat{\sigma}\) is the neutron spin operator, \(\hat{T}\) the neutron nuclear interaction operator and \(\hat{M}_Q\) the neutron magnetic interaction operator, the latter is subscripted \(Q\) to indicate that it only has components perpendicular to \(Q\). We now simplify Eq. 26 by first assuming that there is no alignment of nuclear spins and then neglecting all terms which contribute to incoherent scattering only. The remaining neutron nuclear interaction is then just described by a scalar scattering length \(b\) which is the same at every site. The two intensities measured are given by the cross-sections \(\sigma^{++} + \sigma^{-+}\) and \(\sigma^{--} + \sigma^{+-}\). In the experiment \(Q\) is confined to the horizontal plane and is perpendicular to the magnetisation. It is convenient to define axes \(z\) parallel to the magnetisation direction \(y\) parallel to \(Q\) and \(x\) completing the right-handed
orthogonal set. Then

\[ \sigma^{++} \propto \sum_{il'} \sum_{\lambda\lambda'} p_{\lambda \lambda'} \langle \lambda | \exp[-iQ \cdot R_i(b + M_{zt})] | \lambda' \rangle^2 \]  

(27)

\[ \sigma^{--} \propto \sum_{il'} \sum_{\lambda\lambda'} p_{\lambda \lambda'} \langle \lambda | \exp[-iQ \cdot R_i(b - M_{zt})] | \lambda' \rangle^2 \]  

(28)

\[ \sigma^{-+} \propto \sum_{il'} \sum_{\lambda\lambda'} p_{\lambda \lambda'} \langle \lambda | \exp[-iQ \cdot R_i M_{zt}] | \lambda' \rangle^2 \]  

(29)

and \( \sigma^{+-} = \sigma^{-+} \). The cross-section of Eq. 27 expands to give four terms each of which can be expressed in terms of thermal averages as in section 2. The polarisation dependent terms are those containing products of magnetic and nuclear interactions, they have the same magnitude but opposite signs in \( \sigma^{++} \) and \( \sigma^{--} \). They may be written

\[ \left( \frac{d^2 \sigma}{d\Omega dE'} \right)_{pol} = \int_{-\infty}^{\infty} \left( \left\langle b \exp[-iQ \cdot \hat{R}_{\tau}(0)] \exp[iQ \cdot \hat{R}_i(t) \hat{M}_{zt}] \right\rangle 
+ \left\langle \exp[-iQ \cdot \hat{R}_{\tau}(0)] \hat{M}_{zt} b \exp[iQ \cdot \hat{R}_i(t)] \right\rangle \right) dt \]  

(30)

This polarisation dependent part of the cross-section can be factorised into space and spin dependent parts in the same way and with the same assumptions as can the purely magnetic cross-section (section 2) and gives rise to polarisation dependent magneto-vibrational scattering. The cross-sections \( \sigma^{+-} \) and \( \sigma^{-+} \) are zero for this type of scattering which is elastic in the spin system, so long as the magnetisation is perpendicular to \( Q \). Eq. 30 can be evaluated using the assumptions and methods of section 4 as

\[ \left( \frac{d^2 \sigma}{d\Omega dE'} \right)_{pol} = \left( \frac{\hbar}{2m_a N} \right) bM_{0 \perp} \exp[-(W + W')] \sum_s \frac{1}{\omega_s} \right. 
\times \int_{-\infty}^{\infty} \left( Q \cdot \epsilon_s (Q \cdot \epsilon_s + \Xi_s) \exp[-i(q \cdot l - \omega_st)] (n_s + 1) 
+ Q \cdot \epsilon_s (Q \cdot \epsilon_s - \Xi_s) \exp[i(q \cdot l - \omega_st)] (n_s) \right) dt \]  

(31)

The ratio between the two intensities measured with momentum transfer \( Q = \tau - q \) and energy loss \( \hbar \omega_s \) corresponding to a phonon mode with wave vector \( q \) frequency \( \omega_s \) and polarisation \( \epsilon_s \) is therefore

\[ \frac{\sigma^{++}}{\sigma^{--}} = \frac{1 + 2\gamma + \gamma^2}{1 - 2\gamma + \gamma^2} \]  

(32)

with

\[ \gamma = \frac{M_{0 \perp}}{b} \exp[W - W'] \left( \frac{Q \cdot \epsilon_s + \Xi_s}{Q \cdot \epsilon_s} \right) \]  

(33)

5.2 Experimental considerations

Figure 1 shows a set of typical dispersion curves for a fcc ferromagnetic metal along
the symmetry directions $\Delta$ (100) and $\Sigma$ (110). In general the magnon branch which is parabolic at low $q$ will cross the phonon branches, which are linear, at some finite energy and in the vicinity of this cross-over the two will not be well resolved. Typical resolution ellipsoids which might be obtained with the triple axis polarised neutron spectrometer IN20 at ILL with a Cu 200 monochromator, a Heusler 111 (polarising) analyser and collimation starting from the source of 30' 40' 40' 40' are indicated. With this shape of ellipsoid constant energy scans which cross both the magnon and phonon wave-vectors will show relatively sharp peaks. These peaks can be fitted using Gaussian line shapes to obtain the positions, intensities and widths of the magnon and phonon peaks superposed on a uniform background. The spectra obtained for opposite final spin states can be analysed together and the positions and widths of corresponding peaks constrained to be equal. The quantity $\gamma$ defined in Eq. 33 which gives the ratio of magnetic to nuclear scattering is calculated from the ratio of the heights of the phonon peaks for opposite spin states using Eq. 32 modified to include corrections for incomplete polarisation and spin reversal.

$$I^+ / I^- = \frac{1 + 2P\gamma + \gamma^2}{1 - 2Pe\gamma + \gamma^2}$$

where $P$ is the polarisation and $e$ the flipper efficiency.

6. Polarisation dependence of the phonon cross-section in Fe$_{65}$Ni$_{35}$

Experiments using the technique described above were carried out on a single crystal of Fe$_{65}$Ni$_{35}$[10]. The crystal was in the form of a cube 12 $\times$ 12 $\times$ 12 mm with edges parallel to [110], [001] and [110], it was mounted on IN20 with the [110] edge vertical in a vertical field produced by a pair of superconducting Helmholtz coils. The field at the sample was set to 2T which is sufficient for saturation at 100 K,
the temperature of the experiment. The monochromator, analyser and collimation were as described above and the spectrometer was used in the constant \( k_f \) mode with \( k_f \) either 4.1 or 2.662 Å\(^{-1} \) depending on the energy transfer. A graphite filter was used to eliminate high order contamination of the beam. Dispersion curves for the phonon and magnon modes in the (111) and (001) directions taken from the published work [11,12] are drawn in Fig. 2. They show the magnon mode crossing the LA (111) phonon mode at \( \approx 12 \) meV and the LA (001) phonon mode at \( \approx 7 \) meV. A series of constant energy scans were carried out which are indicated by the shaded oblongs in Figure 2. Those in the (111) directions were based on the 111 reciprocal lattice point and those in the (001) direction on 002. In each scan the scattered intensity was measured at each point first with the flipper on and then with it off. The scattered intensity for both polarisation states obtained in the scans was fitted as indicated above but introducing the additional constraint that the centres of peaks at \( \tau + q \) and \( \tau - q \) in scans at the same energy should be at equal \( q \). When one of the peaks in the \( a \) scan was identified with a magnon it was constrained to have the same peak heights for both polarisation states. These extra constraints were found to improve the determination of the background which is a critical factor in obtaining a correct intensity ratio. The centres of the fitted peaks are plotted as circles (phonons) and triangles (magnon) in Figure 2. In the scans at energies between 7 and 15 meV it was not possible to separate the phonon and magnon intensities, and this severely reduces the amount of information that can be obtained. Unresolved peaks are shown by superposed triangles and circles in Figure 2.

![Figure 2](image)

**Fig. 2.** Dispersion curves for Fe\(_{63}\)Ni\(_{35}\) in the (100) and (111) directions. The magnon mode [12] is shown as the dashed curve and the LA and TA phonon modes [11] as solid curves. The ranges of the constant energy scans [10] are shown as open oblongs and the positions of the peaks of the excitations observed within them as ▲ (magnon) and ● (phonon).

In the absence of spin lattice interaction the ratio

\[
\gamma = \frac{M_\perp}{\bar{b}}
\]

where \( \bar{b} \) is the mean nuclear scattering length per site. The only \( Q \) dependence of \( \gamma \) is through the magnetic form factor \( F(Q) \) (Eq. 7). Fig. 3 shows the magnetic
contribution to the scattering $M_\perp = \gamma \vec{b}$, derived from the scans over the two phonon modes, plotted against $Q$. It can be seen that for the LA 111 phonon there appears to be a reduction in the scattering with increasing $|q|$ which is not observed in the LA 100 phonon. For the LA phonons $\Xi$ is proportional to $q$ so the model developed in section 4 predicts that the intensity should rise on one side of $q=0$ and fall on the other. As for the Debye Waller term $\exp[W - W'] = \exp[\Xi^2]$ it should increase the intensity on either side of $q = 0$ since $\Xi^2$ must be positive.

Fig. 3. The magnetic scattering amplitude observed in the LA 111 (a) and LA 100 phonon modes as a function of $Q$. The dashed line shows the $Q$ dependence expected from the magnetic form factor.

It is quite easy to see qualitatively that these predictions are specific to the simple model (Eq. 7) and that the results are quite sensitive to its specific form. The expression given in Eq. 7 for the renormalisation of the spin corresponds to a model in which the moment decreases with positive strain and increases with negative strain. The greater the phonon amplitude the greater is the momentum fluctuation which is giving rise to the extra scattering: hence the positive Debye-Waller term. The correlation between the sense of the moment fluctuation and the sense of the strain leads to the phase relationship between the normal scattering and the moment fluctuation scattering implicit in the term $Q \cdot \epsilon - \Xi$. The simple model proposed in section 3 presupposes a continuum of states having moments both smaller and larger than that for no strain. As an alternative suppose there were only states with lower moment than the zero strain value and these were excited only for positive strains, then the function giving the moment deviation would be more complex than that of Eq. 8 and in particular would contain additional terms which were even functions of the strain. In principle it is possible to expand any reasonable functional form giving the moment in terms of the strain as a fourier series and to apply the methods of section 4 to it term by term. Unfortunately the algebra becomes rather cumbersome. The effect of terms even in the strain can be seen by taking the simplest even function:

$$S = S_0 \cos(\xi) = \frac{1}{2} S_0 \left( \exp[i\xi] + \exp[-i\xi] \right)$$  \hspace{1cm} (36)
The fourier series has just two terms \( n = \pm 1 \) so there are four terms in the product which gives the thermal average. The important point to note is that in each term \( \gamma_s \) and \( \eta_s \) are imaginary so that the product \( \langle U^2 \rangle + \langle V^2 \rangle \) becomes \( (Q \cdot \epsilon)^2 + \Xi^2 \). It is the same for all four terms. In the sum of \( \langle UV \rangle \) all the terms containing \( \Xi \) cancel out. For this even function therefore the ratio of magnetic to nuclear scattering is given by

\[
\gamma = \frac{M_0}{b} \exp \left( -\frac{W\Xi^2}{(Q \cdot \epsilon)^2} \right) \tag{37}
\]

7. Study of the "forbidden" TA mode

Whilst determining the optimum energies and wavevectors for the polarised neutron experiments described in the previous section an unexpected third peak was found in the spectrum of excitations with wavevectors in the (001) directions. This excitation which has the same dispersion as the transverse acoustic (TA) mode was seen in scans along reciprocal space direction (100) with either [001] or [011] perpendicular to the scattering plane, although in neither case does the polarisation vector for the TA mode have a component parallel to the scattering vector \( (Q_h00) \). This excitation is referred to as the "forbidden" acoustic mode.

![Graph showing wavevector intensity for FeNi alloys](image)

Fig. 4. Scans along (00Q_h) at 100 K with a constant energy transfer of 15meV for (a) Fe_{65}Ni_{35} and (b) Fe_{50}Ni_{50}. Points marked \( \nabla \) are for scattered polarisation parallel and \( \Delta \) antiparallel to the field direction. The peak marked 1 was identified with the magnon mode, that marked 2 with the LA phonon; the peak marked 3 is the "forbidden mode".

The scans which showed this extra mode were repeated under different experimental conditions e.g. by changing the final wavevector but the "forbidden" mode was observed in all experiments. However measurements carried out under identical experimental conditions on a Ni crystal failed to reveal the "forbidden" mode. Figure 4 shows the intensities of the two opposite polarisation states in scans along \( (0,0,Q_h) \) from \( Q_h = 1.48 - 1.96 \) with a constant energy transfer of 15meV for (a) the Fe_{65}Ni_{35} crystal and (b) a crystal of composition Fe_{50}Ni_{50}. The "forbidden"
mode which is labelled (3) is clearly visible in (a) but is at the limit of significance in (b). It appears that the occurrence of this mode is intrinsic to the Invar composition. It has been suggested [13] that the TA 100 mode can be observed in the 100 direction because local orthorhombic distortions of the cubic site symmetry perturb the dynamical matrix elements and allow mixing of the TA and LA modes, but it is difficult to invoke mode mixing which is strong enough to account for its large intensity. Further experiments have therefore been undertaken [14] to obtain more information about this “forbidden” mode. These experiments seek to examine its Q dependence by making measurements in the 1st Brillouin zone.

Fig. 5. The range of energy and momentum transfer accessible with a given scattering angle $\phi$ for (a) An incident energy of 35 meV ($k = 4.1 \text{Å}^{-1}$) and (b) 100 meV ($k = 6.96 \text{Å}^{-1}$). The dispersion of the magnon, and the LA and TA (100) phonon modes are shown as dashed curves.

7.1 Experimental
The conditions under which neutron scattering can take place are determined by the energy and momentum conservation rules:

$$E - E' = \frac{\hbar}{2m} \left( k^2 - k'^2 \right)$$

$$\hbar Q = h(k - k')$$

(38)

(39)

where $E - E'$ is the energy transfer and $k$ and $k'$ are the incident and final neutron wavevectors. Eq. 39 can be rewritten in terms of the scattering angle $\phi$ as

$$Q^2 = k^2 + k'^2 - 2|k||k'|\cos(\phi)$$

(40)

When carrying out triple axis spectroscopy it is usual to fix the magnitude of either $k$ or $k'$ and vary the other in order to provide the energy transfers required. As the energy transfer is increased the momentum transfer increases, and therefore in order to close the scattering triangle, Eq. 40, $\phi$ must also increase. In order to carry out scans at finite energy transfers in the first Brillouin zone it is necessary to increase either $k$ or $k'$. The range of energy and momentum transfer required is
determined by the dispersion of the "forbidden" mode. If Eq. 38 is substituted into Eq. 40 the range of momentum-energy space accessible for given incident neutron wavevector can be determined. The results are shown in Figure 5 for incident neutron energies 35 meV \( (k=4.1 \text{ Å}^{-1}) \) and 100 meV \( (k=6.96 \text{ Å}^{-1}) \) respectively. The dispersion curves for the magnon \( (\Delta=143 \text{ meV Å}^2) \) and the "forbidden" mode \( (24 \text{ meV Å}^{-1}) \) along the [100] direction are superposed. From these figures it is clear that the "forbidden" mode is not observable in the first zone for any scattering angle if the incident energy is only 35 meV which was the maximum used in previous experiments. Increasing the incident energy to 100 meV enables the mode to be observed for energy transfers above 15 meV at a scattering angle of 4 degrees. Experimentally the minimum usable scattering angle is 2.5 degrees. By careful selection of the incident energy it is therefore possible to investigate the dispersion of the "forbidden" mode in the first Brillouin zone for energy transfers greater than 15 meV.

![Diagram](image.png)

Fig. 6. The dispersion of the "forbidden mode" in Fe₆₅Ni₃₅ measured in the 1st Brillouin zone. The solid line represents the dispersion curve determined for the TA (100) phonon mode [11].

The experiment was carried out on the same single crystal of Fe₆₅Ni₃₅ as used in previous neutron experiments [1]. The crystal was mounted with the [110] axis vertical inside a variable temperature cryostat on the triple axis spectrometer IN1 at ILL Grenoble. IN1 is located on the hot source which provides the high energy incident neutrons needed for the experiment. The exterior tail of the cryostat was increased to a diameter of 1.0 m in order to reduce the background scattering at small scattering angles. As in previous experiments the temperature was initially set to 100 K well below the Curie temperature of 550 K but later scans were also carried out at room temperature and above the Curie temperature at 570 K.

The spectrometer was set up with a vertically focusing Cu (200) monochromator and the scattered beam was also analysed using a Cu (200) crystal. In order to work in the first zone close to the straight through position it was necessary to tighten the collimation before and after the specimen to 20'. The spectrometer was operated throughout in the constant \( k' \) mode with \( k' \) set to 5.7 Å\(^{-1}\) for the majority of scans although for some it was necessary to increase \( k' \) to 6.6 Å\(^{-1}\). As
a further check that the "forbidden" mode was intrinsic to the material some scans were repeated with the curvature of the monochromator set to zero i.e. flat, to improve the vertical resolution and, as expected, there was no change in the observed excitations.

An initial longitudinal scan along (001) with an energy transfer of 18 meV showed a strong excitation centred at $q = 0.48$ which was identified with the "forbidden mode". The dispersion of this mode was measured out to the zone boundary and is shown in Figure 6 where it is compared with that of the TA 100 phonon measured by [11]. The constant $Q$ scans from $Q = 0.7$ out to the zone boundary were repeated at 300 K, the dispersion was found to be unchanged, but the intensity was diminished indicating the primarily magnetic character of the scattering. Finally a constant $Q$ scan was carried out at the zone boundary at $T=550$ K ($T_c=500$K) which is compared with that obtained at 100 K in Fig. 7. Only a small fraction of the intensity remains.

These experiments strongly suggest that the origin of the scattering in the "forbidden" mode is not directly due to the displacements of the atoms from their ideal positions but to a secondary effect, such as that suggested in section 2, induced by such displacements. For the former the physics of the scattering process gives an intensity proportional to $Q \cdot \epsilon$ as $q \to 0$ Eq. 7 which is incompatible with the observed behaviour. On the other hand if the scattering is due to magnetic fluctuations driven by the TA100 phonons then, as was shown in section 4, the intensity is proportional to $\Xi^2$, so long as there are odd terms in the dependence of the moment on the strain. For the TA 100 phonon $\Xi$ is proportional to $q$ and to the component $x_{12}$ of the coupling matrix. A dependence on $q$ rather than $Q$ will explain the strength of the scattering in the 1st Brillouin zone, but a non-zero coefficient $x_{12}$ is also necessary. Fig. 8 illustrates how this might arise; it shows the 100 plane of the fcc structure with the displacements corresponding to the TA 100 phonon indicated in an exaggerated way. The perturbation of the crystal field due to the strain is to first order the superposition of a rotation and an orthorhombic distortion. The former couples the $t_{2g}(xy)$ states with the $e_g(x^2 - y^2)$ states and

![Figure 7](image-url)
the latter couples with their spatial extent. Transitions from high moment (HM) to low moment (LM) states might well be induced by this distortion. According to the band structure calculations [3], the LM iron moments on the low moment side of the magneto-volume instability are mostly of $t_2g$ character while HM iron atoms are mostly of $e_g$ character, whereas on the other side of the instability the reverse is true.

![Diagram of strain induced in the nearest neighbour environment of atoms in the 110 plane of an fcc structure by (a) the TA 100 and (b) the TA 110 lattice modes. In (a) the first order strain corresponds a rotation $\alpha$ coupled to an orthorhombic distortion $\delta$, whereas in (b) it is a rotation only.](image)

It is therefore possible that magnetic fluctuations driven by the lattice vibrations can give rise to scattering which has the characteristics of the “forbidden” mode.

8. Conclusions

It has been shown that measurements of the polarisation dependence of the phonon intensity in a ferromagnet can give new information about the interactions between the magnetic and vibrational degrees of freedom. To obtain meaningful results however the phonon and magnon excitations must be sufficiently well resolved, and sufficiently well fitted to ensure that there is no contamination from the magnon in the scattering ascribed to the phonon. This requires better resolution and statistical precision than that needed just to determine the peak positions. Nevertheless polarisation dependent scans made in the region where the magnon and phonon cross, can be used to distinguish the phonon and magnon peaks and to recognise anticrossing behaviour.

In the foregoing sections a very crude model of a possible mechanism of spin-lattice interaction was introduced and some of its consequences for the neutron scattering cross-sections developed. Many physical details have been omitted from this model, including whether the magnetic moment fluctuations which couple to the lattice modes are solely longitudinal. In addition the initial assumption, that the time-scale of the magnetic moment fluctuations is much shorter than that of the lattice vibrations, must break down when the energy difference between the
competing magnetic states becomes comparable with the energy of the phonon which couples them. It is not therefore to be expected that there will be more than qualitative agreement between the predictions of the model and the experiment. Some significant physics does however emerge from the experimental data which any more sophisticated model must contain.

(1) There is an interaction between the LA phonons and the magnetic system which leads to a variation of the ratio of magnetic to nuclear scattering with $q$ additional to that expected from the form factor. This is not present in the TA modes.

(2) The interaction of the magnetic system with the TA (100) phonon leads to a strong scattering cross-section even when the phonon polarisation is perpendicular to the scattering vector ("forbidden mode"). The scattering is probably mainly due to magnetic moment fluctuations driven by, and coupled to the TA 100 phonon mode. No such scattering is observed in the TA 110 mode, suggesting that distortive as well as torsional strain is involved in the coupling.

(3) The polarised neutron data together with the low $Q$ results show that the "forbidden" mode must have a small but significant nuclear contribution. Some mode mixing such as that suggested by [13] must therefore occur.

(4) The exact positions of the phonon and magnon peaks near to the crossover give little evidence for any coupling between the transverse magnetic (spin-wave) excitations and the LA 100 lattice vibrations.

Most of this new information has been obtained from the polarisation dependence of the intensity of the inelastic scattering. One may conclude that when spin-lattice interactions are important it may well be worth taking the extra time needed to measure the polarisation dependence of their inelastic spectra.

9. References

5. O. Halpern and M.H. Johnson Phys. Rev. 55 (1939) 898


