Polarization analysis of the magnetic excitations in Invar Fe$_{86}$B$_{14}$

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Triple-axis polarized inelastic neutron scattering experiments have been carried out on the amorphous ferromagnet Fe$_{86}$B$_{14}$ to separate the longitudinal fluctuations from the transverse (spin wave) excitations. The data suggest that longitudinal excitations exist not only in the vicinity of T_C, but substantially below the ordering temperature as well. The existence of these "hidden" excitations may well explain the "Invar anomaly".

The spin dynamics of isotropic ferromagnets at modest temperatures is known to be well described by linear spin wave theory. In the long wavelength limit there is a Goldstone mode with dispersion relation given by $E_{sw} = D(T)q^2$. The quantitative value of the "stiffness" constant D depends on the details of the interactions and the nature of the magnetism, but the general form of the spin wave dispersion relation, and hence the spin wave density of states, is invariant. The leading order temperature dependence of the magnetization is then given by

$$M(T) = M(0)[1 - BT^{3/2}],\qquad(1)$$

where the coefficient B is related to the spin wave dispersion relation by

$$B = \frac{\zeta(3/2)g\mu_B}{M(0)}\left(\frac{k_B}{4\pi D}\right)^{3/2}.\qquad(2)$$

A measurement of the spin wave dispersion relation can then be directly related to the bulk magnetization, and vice versa.

These relationships, as well as many others provided by spin wave theory, have been found to be in excellent accord with experimental observations for the vast majority of isotropic ferromagnetic materials, with the singular exception of Invar systems [1–4]. In all the Invar materials, whether they be amorphous or crystalline, eq. (2) is found to fail in a major way, with the observed stiffness constant as much as a factor of two larger than that inferred from magnetization measurements. Thus the measured magnetization decreases much more rapidly than can be accounted for based

on the measured dispersion relations. In an attempt to understand the origin of this discrepancy, we previously carried out extensive unpolarized neutron measurements on the amorphous Invar Fe–B system in order to make a detailed comparison between spin wave theory and experiment [3]. We found that conventional spin wave theory worked remarkably well in describing the long wavelength spin dynamics of this Invar alloy system: the dispersion relation was quadratic in q, $D(T)$ obeyed the $T^{5/2}$ Dyson renormalization, and the spin wave linewidths Γ followed the expected q^4T^2 dependence, all within experimental uncertainties. The bulk magnetization also obeyed eq. (1), i.e. it followed the $T^{3/2}$ behavior, but with a calculated stiffness constant which is half the measured value.

The conventional explanation for the observed behavior is that there are additional "hidden" excitations which participate in reducing the magnetization. The magnetization and neutron measurements already put stringent conditions on the form that such excitations might take, since there is no freedom to change the font of the theory, namely the $T^{3/2}$ behavior for the magnetization, the $T^{5/2}$ behavior for $D(T)$, etc. Hence we must have a density of "hidden" excitations which has precisely the same form as the conventional spin wave excitations themselves. One possibility is that the (transverse) spin wave excitations couple to the longitudinal fluctuations, yielding propagating longitudinal excitations which peak at the transverse spin wave energies.

We have been carrying out inelastic polarized neutron measurements on the amorphous Fe$_{86}$B$_{14}$ Invar system to explicitly separate the longitudinal spin fluctuation spectrum (S^z) from the usual spin wave excitations represented by $S^{\pm} = S^x \pm iS^y$. The experiments were carried out on the BT-2 triple-axis polarized beam spectrometer at the National Institute of Stan-

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dards and Technology Research Reactor. Heusler alloy crystals in reflection geometry were employed as polarizing monochromator and analyzer. A pyrolytic graphite filter was used to suppress higher order wavelengths. The collimation was typically $10'-10'-10'-20'$ (FWHM), and the instrumental flipping ratio was ≈ 22 under these conditions. Due to the amorphous nature of the sample, all the present data have been taken in the small wave vector regime. The isotopically enriched $Fe_{86}B_{14}$ sample itself was in the form of stacked ribbons 6 cm long and 0.6 cm wide, and magnetized along the long direction. The Curie temperature was 556 K, and the low T spin stiffness coefficient was ~ 120 meV $Å^2$ [3].

The polarization analysis technique as applied to this problem is in principle straightforward [5]. All the transverse spin wave scattering, represented in the Hamiltonian by the raising and lowering operators $S^{\mp}$, causes a reversal of the neutron spin. These spin-flip cross-sections are denoted by $(+-)$ and $(-+)$. If the neutron polarization $\hat{P}$ is parallel to the momentum transfer Q, $\hat{P} \| Q$, then we may create a spin wave in the $(-+)$ configuration, or destroy a spin wave in the $(+-)$ configuration. Longitudinal fluctuations, on the other hand, are invisible when $\hat{P} \| Q$. Figure 1 shows a measurement with the $(+-)$ configuration. The strong peak on the energy gain side $(E < 0)$ corresponds to the destruction of spin wave excitations. The weak peak on the energy loss side $(E > 0)$ is caused by the imperfect polarization and the fact that $\hat{P}$ was not precisely parallel to Q for this particular measurement.

In the configuration where $\hat{P} \perp Q$, the spin wave scattering still causes a neutron spin-flip, but the energy gain and energy loss cross-sections are now equal (aside from the detailed balance factor), with $\frac{1}{4}$ the intensity of the $\hat{P} \| Q$ configuration. The non-spin-flip
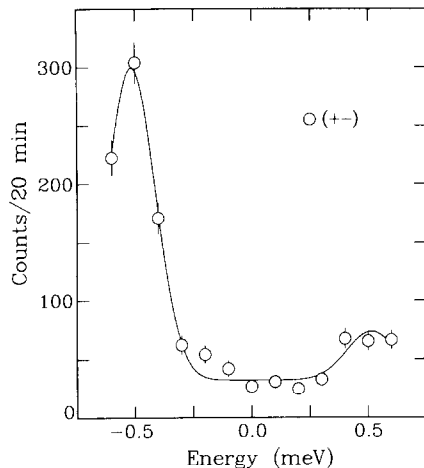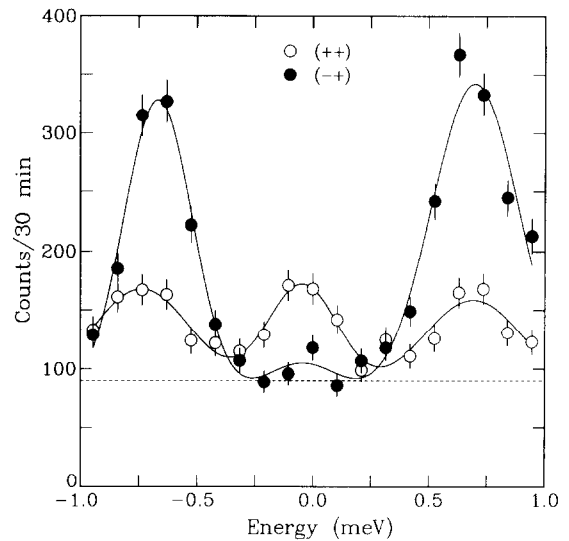

Fig. 2. Observed scattering for $q = 0.09$ Å^{-1} and 465 K in the vertical field configuration $(\hat{P} \perp Q)$. The spin-flip scattering exhibits the usual spin wave excitations, while the non-spin-flip scattering also reveals excitations near the spin wave energies.

$(++)$ or $(--)$ scattering, on the other hand, is directly related to the longitudinal (S^z) scattering. Figure 2 shows a measurement in this vertical field configuration. The spin-flip scattering clearly shows spin waves in energy gain and energy loss, as expected. The non-spin-flip data, on the other hand, also display peaks at the spin wave energies. There is also a peak at $E = 0$, which originates from nuclear scattering. The scattering at the spin wave positions is $\sim 1/3$ the strength of the spin-flip scattering, while the flipping ratio is ~ 10. We make the following remarks about this non-spin-flip scattering: (1) the peak in energy obeys a q^2 dependence; (2) the ratio of the intensity of the spin-flip to non-spin-flip scattering does not change when experimental improvements doubled the flipping ratio; (3) the ratio did not change significantly as a function of q, while the resolution effects [6] change substantially.

These data strongly suggest that there are longitudinal propagating excitations in this Invar system, which appear close to the spin wave excitation energies. These are just the type of excitations which would be needed to explain the "Invar anomaly". However, a word of caution is in order, as the longitudinal and transverse excitation energies are very close to each other, and this is the most likely situation under which "spurious" longitudinal scattering, such as due to the finite flipping ratio, misalignment of $\hat{P}$ and Q, etc., might be found. Therefore further experimentation is warranted before unambiguous conclusions can be drawn. Further work is in progress.

Fig. 1. The $(+-)$ spin-flip scattering cross-section for $\hat{P} \| Q$ observed at 295 K and $q = 0.06$ Å^{-1}. The energy gain side shows a strong spin wave excitation.

Acknowledgement

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