Polarization analysis of the magnetic excitations in Invar and non-Invar amorphous alloys

J. W. Lynn and N. Rosov
Reactor Radiation Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899 and Department of Physics, University of Maryland, College Park, Maryland 20742

G. Fish
Corporate Research, Allied Signal, Inc., Morristown, New Jersey 07962

Conventional spin wave theory works remarkably well in describing the spin dynamics of both Invar and non-Invar isotropic ferromagnets, with the important exception that for Invar systems the magnetization decreases much more rapidly with temperature than can be explained based on the measured spin wave dispersion relations. We have been carrying out triple-axis polarized inelastic neutron scattering experiments on the amorphous ferromagnets $Fe_{86}B_{14}$ (Invar system) and $Fe_{40}Ni_{40}P_{14}B_6$ (METGLAS® 2826) in order to separate the longitudinal magnetic fluctuations from the transverse (spin wave) excitations, and thereby determine if the presence of longitudinal excitations might resolve this discrepancy. The present measurements exhibit longitudinal excitations below T_c, but in both materials. Possible interpretations of the results are discussed.

In the long wavelength (small q) regime the spin wave dispersion relation for an isotropic ferromagnet is given by $E_{sw}=D(T)q^2$, where D is the spin wave "stiffness" constant (Ref. 1). The general form of the spin wave dispersion relation, and hence the spin wave density of states, is the same for all isotropic ferromagnets, while the numerical value of D depends on the details of the magnetic interactions and the nature of the magnetism. The leading order temperature dependence to the magnetization is then given by $M(T)=M(0)[1-BT^{3/2}]$, where the coefficient B is related to the spin wave dispersion relation by

$$B=\frac{2.612g\mu_B}{M(0)}\left(\frac{k_B}{4\pi D}\right)^{3/2}. \qquad (1)$$

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.[2–5] In all the Invar materials, whether they be amorphous or crystalline, Eq. (1) is found to fail in a major way, with the observed stiffness constant as much as a factor of 2 larger than inferred from magnetization measurements. In particular, 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.[4] We found that conventional spin wave theory worked remarkably well in describing the long wavelength spin dynamics of this system, and thus these unpolarized neutron measurements did not suggest an answer to this problem.

The conventional explanation for this "Invar anomaly" is that there are additional "hidden" excitations which participate in reducing the magnetization. If this explanation is correct, then 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 *form* of the theory, viz. 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 which has been suggested[6] is that the (transverse) spin wave excitations couple to the longitudinal fluctuations, yielding propagating longitudinal excitations which peak at the transverse spin wave energies. In an unpolarized beam experiment, such transverse and longitudinal excitations cannot be distinguished. We therefore have been carrying out inelastic polarized neutron measurements on the $Fe_{86}B_{14}$ 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 measurements reveal the presence of longitudinal excitations, not only in the vicinity of T_c,[7] but substantially below the ordering temperature as well.[8]

The experiments were carried out on the BT-2 triple-axis polarized beam spectrometer at the National Institute of Standards and Technology Research Reactor. Heusler alloy crystals in reflection geometry were employed for both monochromator/analyzer and polarizers. A pyrolytic graphite filter was used to suppress higher order wavelengths. Due to the amorphous nature of the sample, all the present data have been taken in the small wave vector (small angle) regime, where the spin waves are well defined at low temperatures. In this regime tight collimation must be employed, and typically we used 10′-10′-10′-20′ (FWHM) in these experiments. The $Fe_{86}B_{14}$ sample itself was in the form of stacked ribbons 7.5-cm long and 0.3-cm wide, and magnetized along the long direction. The neutron beamwidth was restricted to 1.6 cm to suppress edge effects. The flipping ratio measured through the ferromagnetic sample was between 5 and 10, depending on the temperature and experimental setup. The Curie temperature for this material is 556 K, and the low T spin stiffness coefficient is ~ 120 meV Å^2.[4]

The polarization analysis technique as applied to this

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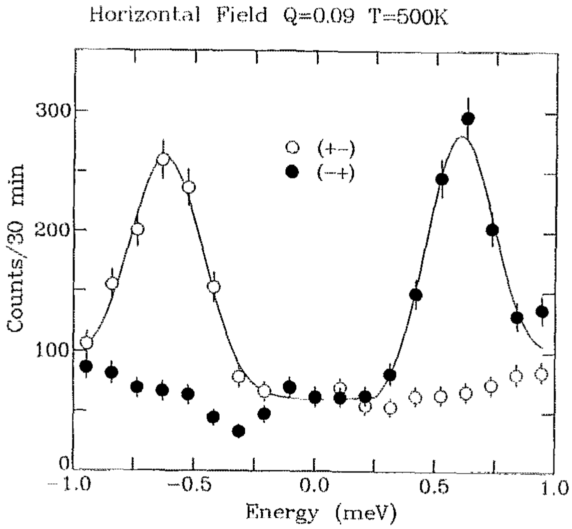

FIG. 1. Spin-flip scattering observed for the amorphous Invar $Fe_{86}B_{14}$ system in the $\hat{P}\|$ **Q** configuration. Spin waves are observed for neutron energy gain ($E<0$) in the (+ −) cross section, and for neutron energy loss ($E>0$) in the (− +) configuration.

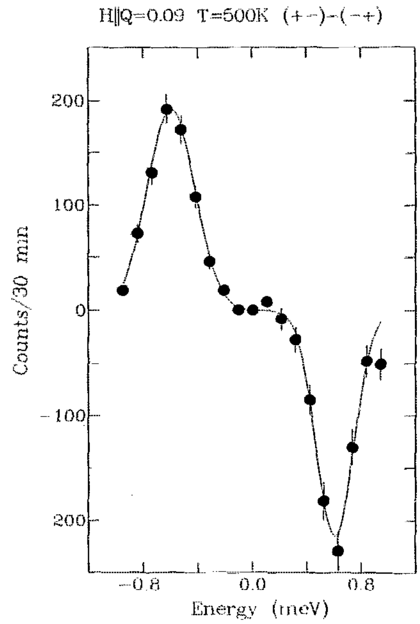

FIG. 2. Subtraction of the measured (− +) data from the data obtained with the (+ −) configuration. The background cancels completely, and only the inelastic magnetic spin-flip scattering survives. This subtraction technique emphasizes the antisymmetric nature of the dynamic susceptibility.

problem is, in principle, straightforward.[9] The (transverse) spin wave scattering, represented in the Hamiltonian by the raising and lowering operators $S^{\pm}=S^x \pm iS^y$, 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 ($E>0$) in the $(-+)$ configuration, or destroy a spin wave ($E<0$) in the $(+-)$. Figure 1 shows measurements in this configuration for $Fe_{86}B_{14}$. We have chosen a temperature of 500 K in order to soften the spin waves so that they are in a convenient energy range to measure. Note that for the $(-+)$ configuration the spin waves can only be observed for neutron energy loss scattering ($E>0$), while for the $(+-)$ configuration spin waves can only be observed in neutron energy gain ($E<0$). We remark that at this wave vector the scan is restricted in energy to ± 1 meV due to kinematic constraints.[2]

In the energy range opposite the spin wave peak in each of the spin-flip configurations in Fig. 1, there is no evidence of a peak in the incorrect configuration, which indicates that the instrument is working well. We do note, however, that there is a small variation in the background with energy. In this case this variation is caused by the rotation of the sample in the beam to maintain the $\hat{P}\|$ **Q** condition, which effectively changes the thickness of the sample in the beam. We do not observe this energy-dependent background in the vertical-field configuration which will be discussed below, as in that situation it is not necessary to rotate the sample. In the horizontal field case one way to eliminate background considerations is to subtract the $(-+)$ cross section from the $(+-)$ cross section, and such a subtraction is shown in Fig. 2. The background scattering cancels, leaving only the spin-flip scattering. We remark that this scattering can be directly related to the dynamic susceptibility $\chi(\mathbf{q},\omega)$, and this subtraction procedure emphasizes the fact that $\chi(\mathbf{q},\omega)$ is an antisymmetric function of ω.

To observe possible longitudinal magnetic fluctuations, we need the experimental configuration where $\hat{P}\perp$ **Q**. In this case the spin wave scattering still causes a neutron spin flip, but it shows up with equal intensity in the energy gain and energy loss cross sections, with 1/4 the intensity of the $\hat{P}\|$ **Q** configuration. Hence the $(+-)$ and the $(-+)$ cross sections are equal. The non-spin flip $(++)$ or $(--)$ scattering, on the other hand, is directly related to the longitudinal (S^z) scattering. Figure 3 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 near the spin wave energies. There is also a peak at $E=0$, which originates from nuclear scattering and elastic magnetic disorder 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, and at a given q is shifted to somewhat higher energy than the spin-flip scattering. (2) The ratio of the intensity of the spin-flip to non-spin-flip scattering did not change significantly when experimental improvements doubled the flipping ratio. (3) The ratio did not change significantly as a function of q, while the resolution effects[10] 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

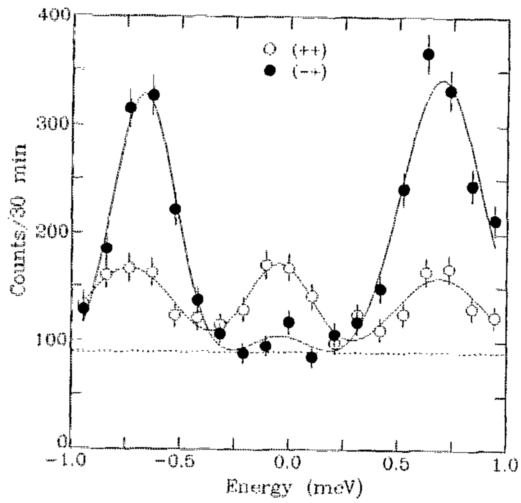

FIG. 3. Observed scattering for the amorphous Invar $Fe_{86}B_{14}$ system in the vertical field configuration ($\hat{P} \perp$ **Q**). The spin-flip scattering exhibits the usual spin wave excitations, while the non-spin-flip scattering also reveals peaks near the spin wave energies.

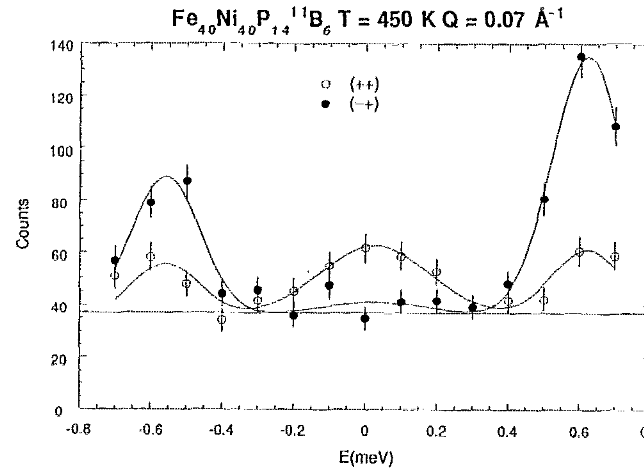

FIG. 4. Spin-flip and non-spin-flip scattering observed for amorphous $Fe_{40}Ni_{40}P_{14}B_6$. The non-spin-flip data also exhibit (weak) peaks at the spin wave energies, but with an intensity which is more than expected based on the measured flipping ratio (see the text).

the type of excitations which would be needed to explain the Invar anomaly. However, it is desirable to determine if these excitations are observed in other amorphous systems which are not Invar materials. We therefore undertook measurements on an amorphous sample of $Fe_{40}Ni_{40}P_{14}B_6$ (METGLAS® 2826), which behaves as a conventional ferromagnet and has been studied in detail.[2,11] Figure 4 shows data for the $\hat{P} \perp$ **Q** configuration. Even though the flipping ratio is only five for these data, and overall the data differ substantially in important details compared with the longitudinal scattering observed in the $Fe_{86}B_{14}$ Invar material, the non-spin-flip "spin wave" peaks are stronger in intensity than expected based on the measured flipping ratio. The extra intensity might be due to longitudinal fluctuations which are expected in all isotropic ferromagnets sufficiently close to T_c (T_c=510 K in this case), and one of the objectives of our polarized beam studies is to investigate this type of scattering as well. Of course the same argument might be advanced for the $Fe_{86}B_{14}$ since both sets of measurements have been taken at elevated temperatures in order to move the spin waves into an experimentally accessible energy range. However, for the $Fe_{86}B_{14}$ system we observe these longitudinal excitations at substantially smaller reduced temperatures. We also remark that we have taken preliminary polarized beam data on a single crystal of $Fe_{65}Ni_{35}$ (Invar), and we have observed clear evidence of longitudinal fluctuations below T_c.[12] Hence we believe that the longitudinal excitations we have observed in $Fe_{86}B_{14}$ may be related to the Invar anomaly, but clearly further work is warranted.

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