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LETTER TO THE EDITOR

Is iron-rich amorphous Fe–B asperomagnetic?

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Abstract. Polarised neutron scattering experiments have recently indicated that amorphous $\text{Fe}_{83}\text{B}_{17}$ is asperomagnetic at room temperature, with up to 20% of the magnetic moments failing to align in the direction of a 4 T magnetic field. We report Mössbauer measurements of the degree of non-alignment for $\text{Fe}_{86}\text{B}_{14}$ and $\text{Fe}_{80}\text{B}_{20}$ at 295 K and 4.2 K. For fields above 2 T, the angles of non-alignment (α) are such that $\langle \sin^2 \alpha \rangle$ is less than 0.07 for $\text{Fe}_{86}\text{B}_{14}$ and less than 0.05 for $\text{Fe}_{80}\text{B}_{20}$ at both temperatures. Upper limits of 10% for $\text{Fe}_{86}\text{B}_{14}$ and 7% for $\text{Fe}_{80}\text{B}_{20}$ can be set on the proportion of randomly aligned moments.

Recent neutron scattering experiments have led to the conclusion that when melt-spun $\text{Fe}_{83}\text{B}_{17}$ is subjected to a magnetic field of 4 T at room temperature, up to 20% of the magnetic moments fail to align in the applied field direction [1]. The field was parallel to the ribbon plane, and polarisation analysis was performed on both the incident and the scattered beams. It was found that the spin-flip cross-section was non-zero, decreased steadily with increasing wavevector transfer, and exhibited a peak positioned at the peak of the structure factor. It was suggested that the spin directions of almost isolated spins are randomly canted away from the applied field direction, and that there are also some larger canted regions. A model was proposed wherein randomly directed strong crystalline fields pin the magnetic moments.

Supporting evidence was cited from Mössbauer measurements of amorphous $\text{Fe}_{78}\text{B}_{13}\text{Si}_9$ subjected to a transverse magnetic field [2]. Here the relative areas of the $\Delta m = 0$ lines ($A_{2.5}$) were used to derive $\langle \sin^2 \alpha \rangle$, where α is the angle between the γ -ray direction and the magnetisation. Anomalous scatter in the $\langle \sin^2 \alpha \rangle$ values was reported for field strengths comparable to the saturation field of about 0.0002 T, and it was suggested that this may be the result of a non-collinear magnetic arrangement.

The neutron scattering results are interesting since the general view is that the (Fe, Ni)–(B, Si) glasses are magnetically soft, with exchange dominating a very low crystalline anisotropy to produce collinear ferromagnetism [3, 4]. The 4 T field applied in the neutron experiments is much larger than the maximum field of 0.001 T used in [2], and is well above the saturation field.

Systems with anisotropic spin freezing (with random orientations about a preferred axis) are called asperomagnets, and have been reviewed by Coey [5]. There is strong

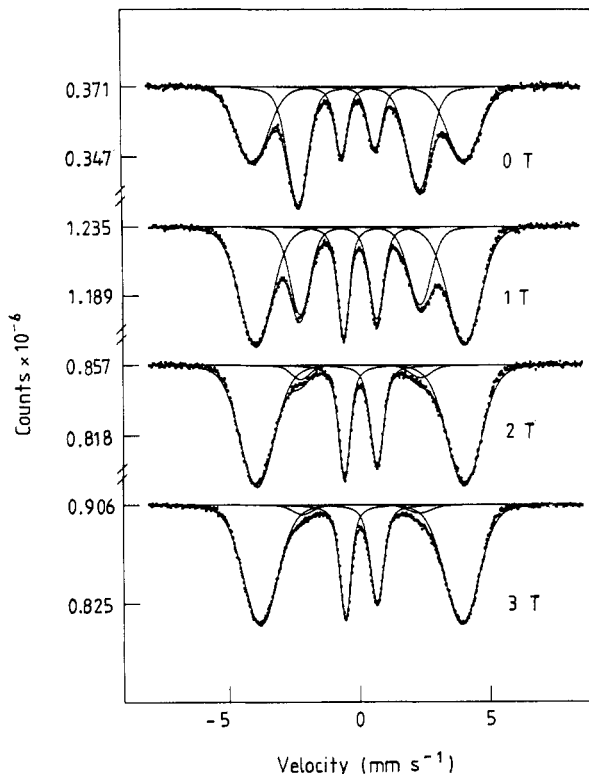


Figure 1. Spectra of Fe₈₆B₁₄ at 295K (RT) subjected to the magnetic fields shown.

evidence that iron-rich Fe–Zr glasses are asperomagnetic because of antiferromagnetic exchange; higher iron concentrations lead to increasingly non-collinear magnetic structures, perhaps as a result of a shortening of Fe–Fe bonds [6]. In Fe–B glasses the boron metalloids fill holes between iron atoms, whereas in Fe–Zr the zirconium atoms are larger than Fe atoms. Consequently, increasingly iron-rich Fe–B and Fe–Zr glasses might predict different polymorphs of pure amorphous iron with different magnetic structures [6, 7].

We performed high-field Mössbauer measurements on Fe₈₆B₁₄ and Fe₈₀B₂₀ glasses at 295 K and 4.2 K to quantify the degree of non-collinearity. The Fe₈₆B₁₄ composition contains almost the highest iron concentration that can be prepared in Fe–B. We might expect non-collinearity to appear at room temperature, as used in the neutron experiments, rather than at 4.2 K, the temperature used for most high-field Mössbauer experiments on metallic glasses.

Ribbons were prepared by melt-spinning under an argon atmosphere. Crystallites of α -Fe and Fe–B were observed in small quantities on the shiny side of as-prepared ribbons using x-ray diffraction and Mössbauer spectroscopy; however, they were removed by gently rubbing the surfaces with fine sandpaper under cyclohexane.

Mössbauer spectra were taken with an 80 mCi ⁵⁷CoRh source moved in a triangular velocity waveform. Spectra with 500 channels were obtained after numerical folding to remove baseline curvature. The samples were mounted in a stress-free holder in a cryostat for all runs; for 4.2 K sample temperatures the source temperature was 4.2 K,

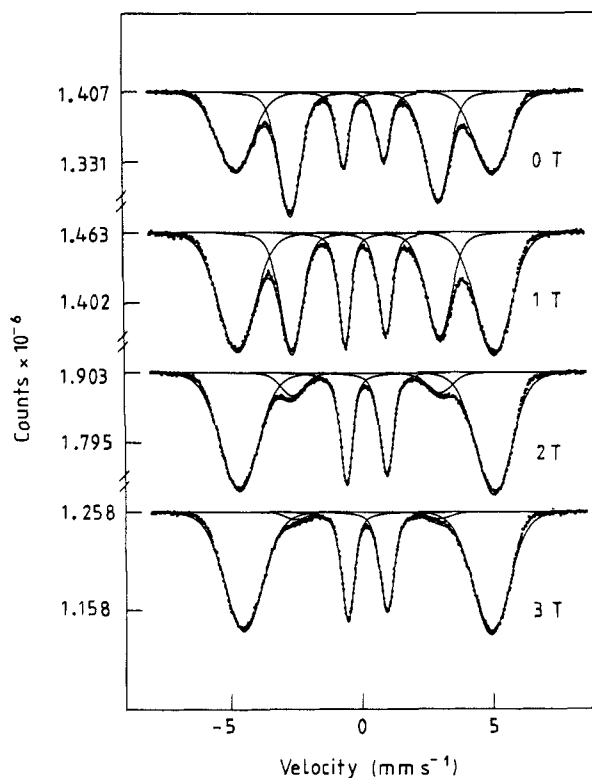


Figure 2. Spectra of $\text{Fe}_{86}\text{B}_{14}$ at 4.2 K subjected to the magnetic fields shown.

whereas for 295 K sample temperatures the source temperature was close to 30 K. Magnetic fields (0.6 T) were applied perpendicular to the plane of the ribbons and parallel to the γ -ray direction. Velocity calibration is with respect to the room temperature spectrum of α -Fe.

Representative Mössbauer spectra taken with increasing applied field strengths are shown in figures 1 and 2. Analysis was performed with the modified Lines and Eibschütz method described elsewhere [8, 9]; the best fits are shown by the full curves in the figures. Pairs of lines were assumed to have equal areas—that is, $A_1 = A_6$, $A_2 = A_5$, and $A_3 = A_4$ —where the lines are numbered in order of increasing valocity. The ratio $A_{25}/A_{34} = 4 \sin^2 \alpha / (1 + \cos^2 \alpha)$, where α is the angle between the γ -ray direction and the magnetic moments, is a measure of the degree of collinearity: for collinear moments it is zero whereas for random moments it is 2. Each spectrum was analysed to measure $\langle A_{25} \rangle / \langle A_{34} \rangle$ and hence derive $\langle \sin^2 \alpha \rangle$.

The broad lineshapes are reasonably well approximated by symmetric profiles, and the spectra were satisfactorily fitted by six Voigtian profiles. The widths and positions of the profiles were constrained to be consistent with the equations deduced from the Hamiltonian for a magnetic hyperfine field (B_{hf}) perturbed by the quadrupole interaction (u) to second order, with distributions allowed for all the hyperfine parameters [9].

There is ambiguity in the interpretation of the applied-field Mössbauer spectra: features that appear to be non-zero A_{25} could in fact belong to A_{16} of a component with a magnetic hyperfine field reduced by the factor $(g_g - g_e)/(g_g - 3g_e) = 0.579$ from that

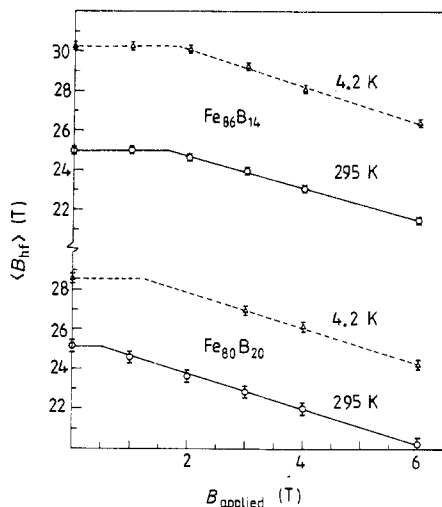


Figure 3. Variation of $\langle B_{\text{hf}} \rangle$ with applied field.

of the main component (a bimodal or skewed magnetic hyperfine-field probability distribution). This is also a problem for $\text{Fe}_{92}\text{Zr}_8$ [10]. Hence a collinear system might appear to have non-zero A_{25} , and consequently the values of A_{25} must be regarded as upper limits.

The distributions for our spectra are not strongly bimodal, since the parameters obtained by fitting (the correlations $\langle \Delta \delta \Delta u \rangle$, $\langle \Delta B_{\text{hf}} \Delta \delta \rangle$, $\langle \Delta B_{\text{hf}} \Delta u \rangle$, and the standard deviations $\sigma(\delta)$, $\sigma(u)$, $\sigma(B_{\text{hf}})$ where δ is the isomer shift) did not change significantly between zero and non-zero applied field spectra, and the positions computed for A_{25} matched the data well. Small differences between the calculated and experimental spectra at extreme velocities indicate that the distributions are not perfectly symmetric, but are somewhat left-skewed.

The variation of $\langle B_{\text{hf}} \rangle$ with applied magnetic field strength is shown in figure 3. For low applied fields $\langle B_{\text{hf}} \rangle$ is approximately constant, whereas for large applied fields a linear dependence is observed. For a collinear ferromagnet subjected to an applied field the hyperfine field is

$$B_{\text{hf}}(B_{\text{applied}}) = B_{\text{hf}}(0) - B_{\text{applied}} + B_{\text{D}} - B_{\text{L}}$$

where B_{D} is the demagnetising field and B_{L} is the Lorentz field. Using the appropriate saturation magnetisations (M_{s}) [11] and calculating the demagnetising factors perpendicular to the ribbon plane, we expect that the term $B_{\text{D}} - B_{\text{L}} \approx \frac{2}{3}\mu_0 M_{\text{s}}$ is 1.0–1.2 T for both samples at both temperatures. This just accounts for the extent of the constant regions in figure 3. In the linearly decreasing regions the slopes of the lines are close to -1 : for $\text{Fe}_{86}\text{B}_{14}$ the slope is $-0.80(10)$ at 295 K and $-0.92(8)$ at 4.2 K; for $\text{Fe}_{80}\text{B}_{20}$ it is $-0.89(14)$ at 295 K and $-0.90(16)$ at 4.2 K. Hence the majority of the moments are collinear in these regions.

It can be seen from the $\langle A_{25} \rangle / \langle A_{34} \rangle$ values plotted in figure 4 that the data for applied fields greater than 2 T allow a small amount of non-collinearity: $\langle \sin^2 \alpha \rangle$ is less than 0.07 for $\text{Fe}_{86}\text{B}_{14}$ and less than 0.05 for $\text{Fe}_{80}\text{B}_{20}$ at both 295 K and 4.2 K. If any non-collinear moments are assumed to be randomly directed, their relative abundances are less than 10% for $\text{Fe}_{86}\text{B}_{14}$ and less than 7% for $\text{Fe}_{80}\text{B}_{20}$. These amounts are smaller than that (up to 20%) reported from the neutron experiments with a 4 T field applied [1]. Smaller

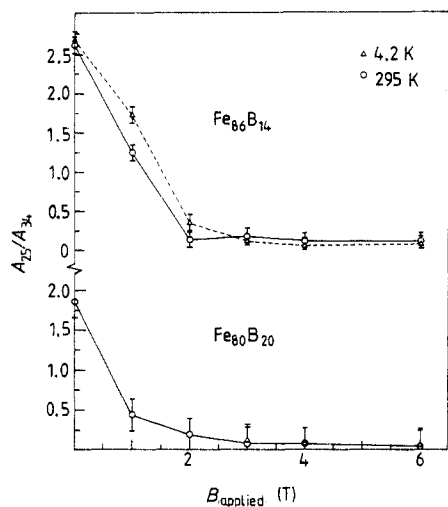


Figure 4. Variation of $\langle A_{25} \rangle / \langle A_{34} \rangle$ with applied field.

upper levels on the proportion of randomly aligned moments, or non-zero lower levels, cannot be set with conventional Mössbauer spectroscopy, since the effects of non-collinearity and the effects of a left-skewed hyperfine-field distribution cannot be distinguished.

Further investigation is required to determine the cause of the anomalous scatter in the Mössbauer line area ratios reported for field strengths comparable to the saturation field [2]. However, a recent study has shown that accurate measurements of Mössbauer line areas can be extremely difficult in cases where one or more of the hyperfine parameters is distributed asymmetrically [12].

It would be worthwhile to extend the polarised neutron scattering experiments described in [1] to a range of temperatures and applied field strengths. Evidence supporting asperomagnetism could then be sought for a well characterised specimen, perhaps using small-angle neutron scattering or Mössbauer spectroscopy.

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