NEUTRON SCATTERING STUDY OF THE MAGNETISM IN A NANOCRYSTALLINE/AMORPHOUS MATERIAL

N. ROSOV,* J. W. LYNN,*† AND G. E. FISH**
*Reactor Radiation Division, NIST, Gaithersburg, MD 20899
†Department of Physics, University of Maryland, College Park, MD 20742
** Allied Signal Inc., Morristown, NJ 07962

ABSTRACT

Recently developed nanocrystalline magnetic systems are of considerable interest fundamentally as well as technologically. One such material is $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$, which can be produced by heat treating the amorphous precursor. This forms a noncrystalline phase with typical dimension of 350 Å as determined by neutron diffraction. Small angle neutron scattering (SANS) has been employed to investigate the properties of the nanocrystallized material over the temperature range from 10 K to 725 K, a regime where no significant structural changes are expected to occur. In zero field and low temperature (10 K) we obtained an isotropic scattering pattern. The application of a relatively modest field to sweep out the domains changed the scattering to a "butterfly wings" pattern typical of patterns dominated by magnetic elastic intensity. Up to 450 K this pattern changed only modestly, while for substantially higher temperatures the ratio of inelastic to elastic scattering increased rapidly as the magnetic phase transition of the intergranular component ($\simeq$ 575 K) was approached. Triple axis inelastic measurements showed that the majority of the magnetic inelastic scattering was from the nanocrystalline phase.

INTRODUCTION

Soft amorphous ferromagnets are of great technological importance; however, their application is limited by the recrystallization that occurs at higher temperatures. The discovery of the "so-called" nanocrystalline-amorphous alloys,[1] which maintain extremely soft magnetic properties up to much higher temperatures without recrystallizing, is therefore of great interest. These alloys are produced by controlled recrystallization of an amorphous alloy that produces a two-phase material: a nanocrystalline phase that is embedded in an amorphous matrix. The nanocrystalline and amorphous phases have very different ordering temperatures: for the amorphous phase $T_{Ca} \approx 575$ K, and the nanocrystalline phase remains ordered up to $T_{Cn} \approx 825$ K. Above T_{Ca}, the coercivity increases dramatically, suggesting that the loss of magnetic ordering in the amorphous matrix causes the nanocrystallites to be magnetically decoupled. The maintenance of the soft magnetic properties above the ordering temperature is attributed to the random averaging of the nanocrystalline particles that are smaller than the characteristic length scale for ferromagnetic exchange.[2] Although the static nature of the two-phase coexistence has been elucidated, the dynamic properties of these materials, particularly the magnetic excitations, are not well understood. In this paper, we report a series of neutron scattering measurements on nanocrystalline amorphous $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$.

SAMPLE PREPARATION AND CHARACTERIZATION

Amorphous ribbons were prepared from pre-alloyed ingots of $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$ by planar flow casting onto a copper substrate in an argon atmosphere. The nanocrystalline phase

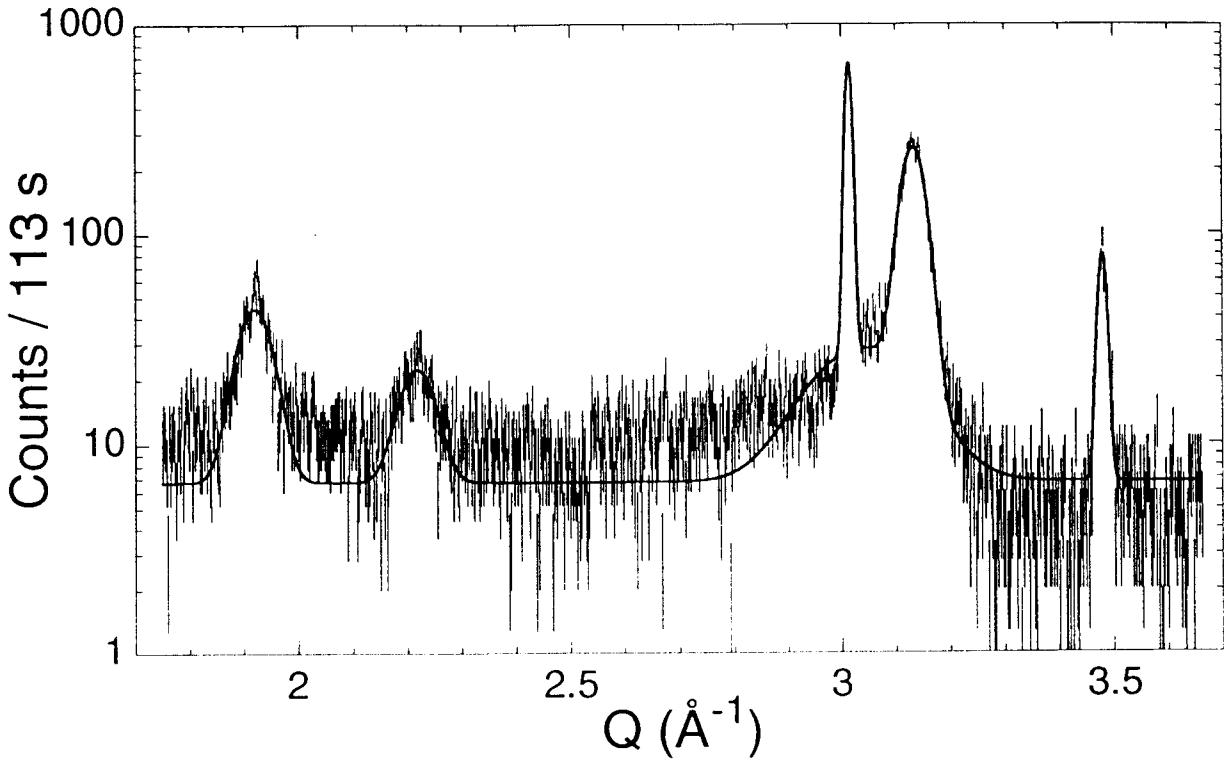

Figure 1: Intensity *vs.* Q for elastic scattering from $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$ at $T = 340$ K. The narrow, resolution-limited peaks are from the Cu sample holder. The broader peaks are due to Fe-Si nanocrystals, and the width implies a particle size of ≈ 350 Å.

was formed by heating the ribbons to 835 K. The sample was composed of 5 cm long strips stacked together and mounted in an open copper frame. The opening was covered by 0.6 mm Cu foil as a heat shield to improve the thermal uniformity of the sample. The sample was mounted in a closed-cycle He refrigerator for $T \leq 350$ K, and in a resistively-heated vacuum furnace for temperatures up to 730 K. The magnetic field, aligned along the long dimension of the ribbons, was produced by an electromagnet.

To characterize the sample, we first performed a diffraction scan on BT-9, a triple-axis spectrometer at the NIST research reactor (NBSR). The $\lambda = 2.44$ Å incident neutrons were produced from the 002 reflections of a pyrolytic graphite (PG) monochromator. The collimation was $10' - 11' -$ sample $- 12' - 16'$, before and after the monochromator and before and after the the PG002 analyzer, respectively. The 340 K diffraction pattern (Fig. 1) shows two set of peaks: The narrow (resolution-limited) peaks are located at 3.013 and 3.480 Å^{-1}; and the broader peaks (which are more than five times broader than the instrumental resolution) are located at 1.92, 2.218, and 3.13 Å^{-1}. There is also an extremely broad component centered at 3.05 Å^{-1} that is ≈ 15 times the instrumental resolution. The narrow peaks can be attributed to the Cu sample sample holder. The broader peaks are indicative of a nanocrystalline phase, and the positions indicate that the nanocrystalline phase is body centered cubic with $a_0 = 2.835$ Å. The presence of the superlattice peaks indicates that the alloy is atomically ordered. The lattice constant is a little larger than that expected for $Fe_{80}Si_{20}$, consistent with the possibility that there is some small admixture of Nb present in the nanocrystallites.[3] The width of the peaks indicates that the characteristic dimension

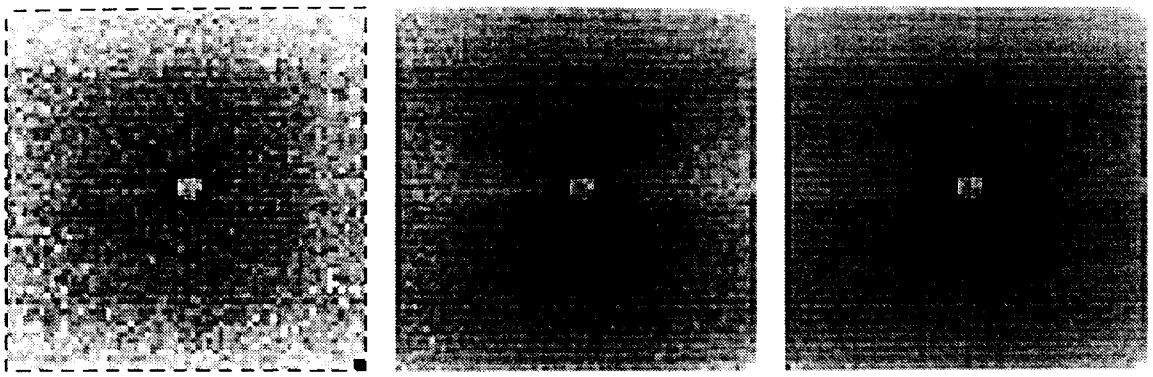

Figure 2: 2D SANS images at various fields and temperatures. The magnetic field is applied horizontally. Zero momentum transfer is at the center of the image, and the x and y limits correspond to $\pm 0.0882\,\text{Å}^{-1}$. (Left) $\mu_0 H_{app} = 0\,\text{T}$ and $T = 10\,\text{K}$; (middle) $\mu_0 H_{app} = 0.1\,\text{T}$ and $T = 10\,\text{K}$; and (right) $\mu_0 H_{app} = 0.1\,\text{T}$ and $T = 725\,\text{K}$.

of the nanocrystalline regions is 350 Å. In addition, about 20% of the scattering is due to an amorphous component.

SANS MEASUREMENTS

In order to observe the small-q scattering from the magnetic microstructure of this nano-amorphous material, we performed small-angle neutron scattering (SANS) measurements on the NG-3 30 m SANS spectrometer at the Cold Neutron Research Facility (CNRF) of the NIST research reactor. A velocity selector produced an incident neutron distribution with a wavelength of 5 Å and a full width at half maximum (FWHM) $\Delta\lambda/\lambda = 15\%$. The detector was 4 m from the sample, four guides were inserted in the incident beam, which gave a minimum accessible scattering vector Q_{min} of 0.0125Å^{-1} and a momentum resolution near the center of the detector of 0.0025Å^{-1}. We started with the sample at $T = 10\,\text{K}$ (see Fig. 2). In the case where there is no applied magnetic field, the magnetic scattering, as well as the structural scattering, is isotropic (Fig. 2). Applying a 1 kOe field drives out all the domains, magnetically saturating the sample, and changes the scattering pattern to an isotropic structural term and two anisotropic magnetic terms. The elastic magnetic scattering has a $1 - \cos^2\phi$ dependence, whereas the inelastic scattering has a $1 + \cos^2\phi$ dependence, where ϕ is the angle around the detector with respect to the direction of the applied field. Thus in the case of a saturated ferromagnet the scattering can be written as a sum of an isotropic term and an anisotropic term:

$$A(q) + B(q)\cos^2\phi = \alpha(q) + \beta(q)(1 + \cos^2\phi) + \gamma(q)(1 - \cos^2\phi) \qquad (1)$$

where the q-dependent terms α, β, and γ are the structural, elastic magnetic and inelastic magnetic scattering, respectively.

In a saturating field at low temperatures, the scattering pattern has a characteristic butterfly pattern, indicating that most of the scattering is elastic; this characteristic pattern persists up to $\approx$ 450K. At the highest temperatures ($\sim$ 700 K), the scattering pattern has changed so that the inelastic term dominates.

TRIPLE AXIS SPECTROSCOPY

For isotropic ferromagnets,[4] the dispersion relation between the spin-wave energy and mo-

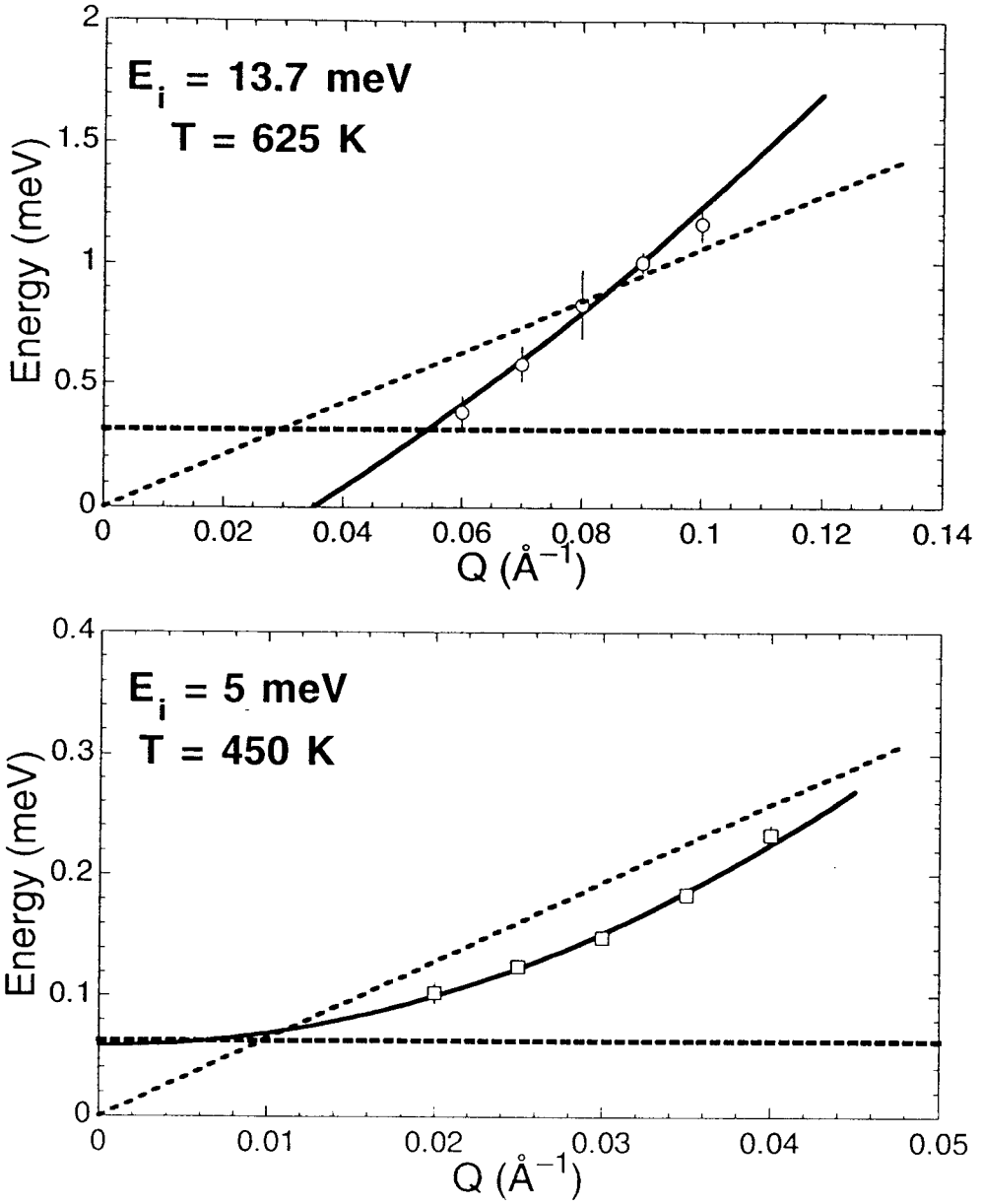

Figure 3: Dispersion curves, E *vs.* q, for nanocrystalline/amorphous $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$. (Top) $T = 625\,K$ and incident energy $E_i = 13.7$ meV. (Bottom) $E_i = 5$ meV and $T = 450$ K. The solid lines are fits of the data to $E = Dq^2 + \Delta$. The dotted horizontal line indicates the energy resolution, while the other dotted line indicates the kinematic limit for these constant-q scans.

mentum is quadratic in the small-q regime, $E = Dq^2$, where D is the spin wave stiffness. We studied the magnetic inelastic cross sections with the BT-2 and BT-9 triple-axis spectrometers at NBSR, in order to determine the source of the inelastic scattering which we observed in the SANS experiments. We performed constant-q, variable energy scans with fixed incident energies of 13.7 meV and 5 meV. The inelastic scans must be made around the (000) position in order to measure the long wavelength spin excitations, since this is the only reciprocal lattice point in an amorphous system. This restricts the spin wave excitations that are experimentally accessible since the maximal achievable energy transfer increases

linearly with moment transfer, $E_{\pm} \approx \pm 2k_i q$, whereas the spin wave energy increases as q^2.

The $E_i = 13.7\,\mathrm{meV}$ scans were performed with collimations of $10'-11'-\mathrm{sample}-12'-16'$ giving an energy resolution at the elastic position of $\approx 0.25\,\mathrm{meV}$, a longitudinal q resolution of $\approx 0.01\,\mathrm{\AA}^{-1}$ in the scattering plane, and a vertical resolution of $\approx 0.22\,\mathrm{\AA}^{-1}$. At the momentum transfers accessible with 0.25 meV energy resolution, the excitations are not at a low enough energy to observe until the sample temperature is greater than $\sim 625\,\mathrm{K}$ (see Fig. 3). The data can be fit to a quadratic dispersion and an energy gap, $E = Dq^2 + \Delta$: at $T = 625\,\mathrm{K}$, the spin wave stiffness is about $125\,\mathrm{meV\AA}^2$. The negative gap is probably an artifact of the observed spin wave energies being so close to the experimental kinematic limit.

We are able to observe spin excitations at lower temperatures by changing the incident energy to 5 meV, thus improving the energy resolution to $\approx 0.06\,\mathrm{meV}$ at the elastic position. We can then observe spin waves over the temperature range $200\,\mathrm{K} < T < 450\,\mathrm{K}$, where the spin stiffness varies from 200 to $150\,\mathrm{meV\AA}^2$.

The large value of the spin wave stiffness and the temperature dependence of the spin wave stiffness indicates that we are observing only the excitations in the Fe-Si nanocrystallites, and not in the amorphous matrix. We, of course, might expect only to observe such excitations as the majority of the material is in the nanocrystallites.

CONCLUSIONS

We have performed a variety of neutron scattering measurements on a nanocrystalline/amorphous material, $Fe_{73.5}B_9Si_{13.5}Cu_1Nb_3$, to characterize the sample, and determine some of the dynamical magnetic properties of the material. Neutron diffraction measurements showed that the preparatory heat treatment produced a sample in which $\approx 80\%$ of the sample was in nanocrystallites with an average size of about 350 Å; the rest of the sample is amorphous. SANS measurements in a magnetic field showed that the inelastic scattering increases with increasing temperature, and that most of the low temperature scattering is elastic magnetic scattering. Constant-q variable energy scans of the magnetic excitations around 000 showed that the observed spin waves were mainly due to the component with the higher Curie temperature, with a rather large spin stiffness at low temperature, as expected for Fe-Si nanocrystallites. Further work will involve the study of microscopic kinetics of the recrystallization process by SANS, as well as a search for the amorphous excitations in an alloy with a greater amount of amorphous phase.

ACKNOWLEDGMENTS

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