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Magnetic Field Dependence of the Small-Angle Neutron Scattering (SANS) in Amorphous DyCu and NdFe₂

Donna Marie Andrauskas
University of Rhode Island

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MAGNETIC FIELD DEPENDENCE OF THE SMALL-ANGLE
NEUTRON SCATTERING (SANS) IN
AMORPHOUS DyCu AND NdFe₂

BY
DONNA MARIE ANDRAUSKAS

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF SCIENCE
IN
PHYSICS

THESIS COMMITTEE

Major Professor

Stephen P. Richard
S. P. Richard

A. C. Nease
A. C. Nease

TEAM OF THE GRADUATE SCHOOL

UNIVERSITY OF RHODE ISLAND

1985

ABSTRACT

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OF

DONNA MARIE ANDRAUSKAS

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APPROVED:

Thesis Committee

Major Professor

Stanley Dickart
S. Milon

A. C. Nunes

A. A. Michel

DEAN OF THE GRADUATE SCHOOL

ABSTRACT

Previous data on amorphous NdFe_2 have shown that it has some sort of phase transition at approximately 330 K. Previous magnetization studies on amorphous DyCu have shown that this alloy undergoes some sort of phase transition at approximately 17 K. Unusual field cooling and time relaxation effects were also observed in these measurements on DyCu below this temperature. Both the exact nature of these phase transitions and the resultant magnetic structures of these alloys are not yet fully understood. This study was undertaken to obtain additional information about them.

Small-angle neutron scattering data on amorphous NdFe_2 were taken in zero field and in applied fields up to 18 kOe. In zero field, the lineshape of the scattering was Lorentzian above the transition temperature and became increasingly deviated from this form below this temperature. The spin correlation length showed a broad maximum near to, but lower than, the transition temperature and then decreased below this temperature. SANS data obtained with the applied field showed a sharp decrease in intensity with increasing field. The spin correlation length also decreased with increasing field.

The response of the SANS from amorphous DyCu to applied fields up to 15 kOe was examined. Application of the field caused a decrease in SANS intensity with

increasing field and also caused a decrease in the correlation length which was obtained from the lineshapes which were Lorentzian to 6 \AA from the zero field value of 13.5 \AA . Time relaxation effects were observed in the field-cooled data.

The SANS measurements showed that amorphous DyCu and NdFe₂ do not have a magnetic phase transition to long-range ferromagnetism. They indicated that these alloys undergo a transition to a state that has some features of a spin glass.

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ACKNOWLEDGEMENT

The work at the University of Rhode Island was supported by National Science Foundation Grant Number DMR8207078. The amorphous DyCu sample was fabricated at IBM through the Industry-University Cooperative Program with IBM. The amorphous NdFe₂ sample was prepared at Battelle Pacific Northwest Laboratories. The SANS measurements were obtained with the area detector small angle spectrometer located at the National Bureau of Standards Reactor. I worked with J.J. Rhyne and H.A. Alperin of NBS and S.J. Pickart and S. Hasanain in obtaining and analyzing the data.

This manuscript is lovingly dedicated to my parents, John and Helen Andrauskas, and my grandmother, Lottie Majewski. Their love, belief in me, and support fueled my courage and determination. For this, I am grateful.

I wish to thank Dr. S.J. Pickart for all the support and assistance that he has given me throughout this period.

I also wish to thank my friends for their loving patience, understanding and encouragement. Also, they were the source of many pleasant and necessary diversions. Thanks for helping me preserve my sanity !

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This master's thesis is written in the manuscript plan format. In this format, the thesis is divided into two sections. Section One contains two papers that have been published in the Journal of Applied Physics. Section Two consists of appendices that contain background information on the papers.

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APPLIED FIELD RESPONSE OF THE SPIN GLASS STATE
IN AMORPHOUS NdFe_2

M.L. Spano and H.A. Alperin

Naval Surface Weapons Center, Silver Spring,
Maryland, 20910

and

National Bureau of Standards,
Gaithersburg, Maryland 20899

J.J. Rhyne

National Bureau of Standards,
Gaithersburg, Maryland 20899

and

S.J. Pickart, S. Hasanain and D. Andrauskas

University of Rhode Island,
Kingston, Rhode Island 02881

ABSTRACT

The magnetic correlations and the response of the spin system to an applied field have been studied in sputtered amorphous NdFe_2 using high-resolution small angle neutron

scattering (SANS) techniques. In zero field the lineshape of the scattering is Lorentzian down to a spin freezing temperature of approximately 330 K. Below this temperature the lineshape becomes increasingly non-Lorentzian and has been analyzed using a Lorentzian plus Lorentzian-squared cross section appropriate for a random anisotropy field system. The correlation length shows a broad maximum of 170 angstroms near 285 K (below T_c) and then decreases to a constant value of about 80 angstroms for $T < 100K$. SANS data taken in applied fields up to 18 kOe at 83 K produced constant intensity contours which are elliptically distorted with the $Q \parallel H$ direction being the major axis. The overall intensity and the calculated spin correlation length both drop abruptly with increased field. This behavior is consistent with the field driving the dominant part of the spin system into an infinite cluster with the residual smaller spin clusters exhibiting a super-paramagnetic type response.

INTRODUCTION

The amorphous rare earth iron alloys exhibit, as a class, a rich variety of magnetic phenomena. Because of the amorphous structure of these materials, the rare earth atoms possess a large random magnetic anisotropy which, according to the theory of Aharony and Pytte [1], profoundly affects the magnetic properties of these alloys. In particular ferromagnetic spin ordering will not occur at any temperature in a system with finite random anisotropy fields, and the correlation length will have a limited range proportional to $(I/D)^2$, where I and D are average values of the exchange and anisotropy parameters.

The focus of this paper is a neutron scattering study of the nature of the phase transition in amorphous NdFe_2 , on its magnetic state below T_c , and on its response to applied fields. Previous magnetization data [2] on this sputtered sample indicated a transition temperature T between 336 K and 365 K, whereas other studies [3,4] on evaporated samples found it to be approximately 380 K. The magnetization curves do not saturate even at fields of 18 kOe. The sharply reduced transition temperature compared to crystalline RFe_2 compounds and the lack of saturation are evidence for the strong local random anisotropy.

Prior neutron scattering experiments [2] on NdFe_2 were performed on a conventional diffractometer and showed a peak in the low Q (Q = wave vector transfer) scattering at

305 K corresponding to the transition temperature. In addition, the neutron data indicated that, as T was reduced to and below T_c , the magnetic correlation length remained finite, indicating clustering, as opposed to long-range ordering, of the spins.

The present data were taken on an area detector small angle scattering spectrometer (SANS) at the NBS Reactor which allowed measurements to smaller Q ($Q_{\min}=0.015 \text{ \AA}^{-1}$) and removed the uncertainty associated with the large corrections for resolution effects in conventional spectrometers.

Small angle neutron scattering data were taken in zero field from 6 K to 410 K and in magnetic fields up to 18 kOe at two temperatures (83 K and 210 K). The qualitative features of the 83 K and 210 K field-dependent data were similar, and only the 83 K results will be presented in this paper. The incident neutron wavelength used in the experiments was 5.5 angstroms, and the range of useful wave vector transfer, Q , was $0.015 \text{ \AA}^{-1} < Q < 0.080 \text{ \AA}^{-1}$. The sample was prepared by rapid dc sputtering onto a cold copper substrate at Batelle Pacific Northwest Laboratories.

SANS RESULTS-ZERO FIELD

Figure 1 shows the neutron intensity as a function of T at various Q 's. The area around the transition

temperature is expanded in the inset. For all Q 's there is a small peak in the intensity at 330 K. This is somewhat higher than the temperature (305 K) of the critical scattering peak in the data of Alperin et al [2]. At lower temperatures an enormous increase in the scattering occurs which reflects the formation of spin clusters as opposed to ferromagnetic order for which the SANS would nearly vanish below T at these Q . Figure 2 plots the inverse intensity versus Q -squared for temperatures both above and below the transition. In a conventional ferromagnet the scattering for $T > T_c$ has a Lorentzian lineshape reflected as straight lines in Figure 2. Note that for $T > T_c$ the lineshape is Lorentzian, but as T is decreased through T_c the data become increasingly curved. We have analyzed this data using a Lorentzian plus Lorentzian-squared cross section which is appropriate for a random anisotropy system, as proposed by Aharony and Pytte [1]. Its form is

$$I(Q) = A/(Q^2 + K^2) + B/(Q^2 + K^2)^2 \quad (1)$$

where K is the inverse of the spin correlation length. The temperature dependences of the parameters A and B are shown in Figure 3. Note that there is a sharp peak in the Lorentzian coefficient at 340 K, which is approximately

the critical temperature as derived from the peak in the scattering intensity. Also observe that, at lower temperatures, the values of both coefficients increase sharply. Values of the spin correlation length as a function of temperature are also given in Figure 3. Note that the correlation length has a broad maximum of 170 angstroms near to, but lower than, the critical temperature. ($T_{MX} \approx 285$ K). Below this temperature the correlation length decreases to approximately 80 angstroms at 100 K, after which it remains constant.

These results may be understood as follows. As T is decreased from above T_c , the alloy appears to approach a conventional ferromagnetic phase transition. This is reflected in the rise in the correlation length as T_c is approached from above. However, below T_c the presence of the random anisotropy on the rare earth sites precludes the correlation of spins beyond a certain length, resulting in clusters of spins. As the temperature is further decreased, the effective local random anisotropy increases, thereby decreasing the correlation length even further. The low T state of this amorphous alloy may be classed as a spin glass.

FIELD DEPENDENCE

In zero field, constant intensity lines on the SANS

area detector appear as circles (isotropic scattering). As a magnetic field is applied to the sample, the scattering becomes elliptically distorted with the major axis in the direction of the field. This prolate distortion is opposite to the oblate distortion expected for a ferromagnet and is characteristic of the response from a system of super-paramagnetic spins or spin clusters. In addition, increasing the applied field sharply reduces the intensity of the scattering. The scattering lineshape in Q , as measured in non-zero applied fields at 83 K, remains non-Lorentzian and has again been fitted to the Lorentzian plus Lorentzian squared cross section (eqn. 1). Due to the anisotropy of the scattering, 30° wide sector averages of the scattered intensity parallel and perpendicular to the H field direction have been individually fit to equation 1. The corresponding correlation lengths $\xi_{Q \parallel H}$ and $\xi_{Q \perp H}$ are plotted in Figure 4 along with the coefficients A and B of the two terms (eqn. 1). It is noted that the correlation length drops rapidly with field to below 20 angstroms for $H > 12$ kOe. In addition there is a relatively strong and anisotropic field dependence to the amplitude coefficients. When the behavior of the coefficients is considered in light of the prolate distortion and overall intensity reduction, the picture emerges of a system of spin clusters of varying size, the larger of which are driven into a near-infinite

ferromagnetic cluster by the application of an external field. Such giant clusters will not contribute to the observed SANS data, since they scatter only infinitesimally close to $Q = 0$. The net effect is that only the smaller clusters remain visible to the SANS, and thus the scattering intensity is reduced. The presence of these loosely coupled, increasingly smaller clusters corresponds to the observed correlation length reduction with increasing field.

CONCLUSIONS

These results plus the previous magnetization data imply that NdFe_2 does not undergo a second order phase transition to a ferromagnetic state, but rather exhibits a spin-frozen cluster state which has a strong sensitivity to applied fields. This is consistent with the prediction of Aharony and Pytte [1] that, in a random anisotropy alloy, no ferro-magnetic phase transition is possible in three dimensions.

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FIGURE 1

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FIGURE 2

Plot of the inverse intensity versus Q^2 for several temperatures. The curves are linear and intersect at Q_0^2 . The solid vertical line is the position of the spin density wave. The variation of the spin temperature with increasing temperature from the Curie temperature (see text).

FIGURE 3

(See text for description of the Curie temperature dependence of the intensity and the position of the spin density wave. The solid vertical line is the position of the spin density wave. The variation of the spin temperature with increasing temperature from the Curie temperature (see text).

FIGURE CAPTIONS

FIGURE 1

Scattered neutron intensity versus temperature in amorphous NdFe_2 for several small Q as given for each curve. The inset shows the weak inflection present at the transition temperature followed by the anomalous increase in scattering at lower temperatures.

FIGURE 2

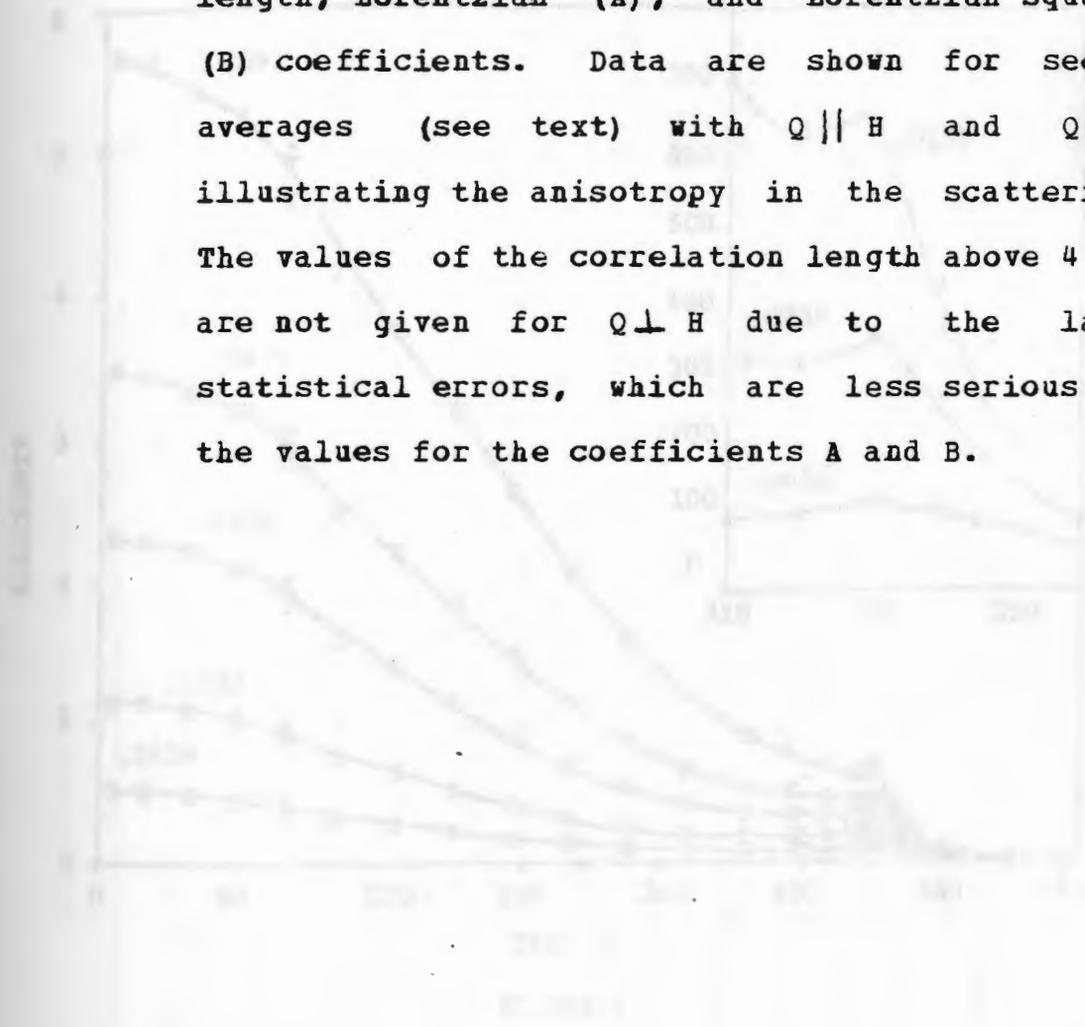
Plot of the inverse intensity versus Q^2 for several temperatures both above and below the transition temperature T_c . The lines represent fitted values of the cross section, and the curvature of the lower temperature data indicates departures from the Lorentzian lineshape (see text).

FIGURE 3

(Top) The temperature dependence of the Lorentzian and Lorentzian squared (A and B of eqn. 1) coefficients. (Bottom) The temperature dependence of the spin correlation length.

FIGURE 4

Applied field dependence of the correlation length, Lorentzian (A), and Lorentzian squared (B) coefficients. Data are shown for sector averages (see text) with $Q \parallel H$ and $Q \perp H$ illustrating the anisotropy in the scattering. The values of the correlation length above 4 kOe are not given for $Q \perp H$ due to the large statistical errors, which are less serious for the values for the coefficients A and B.



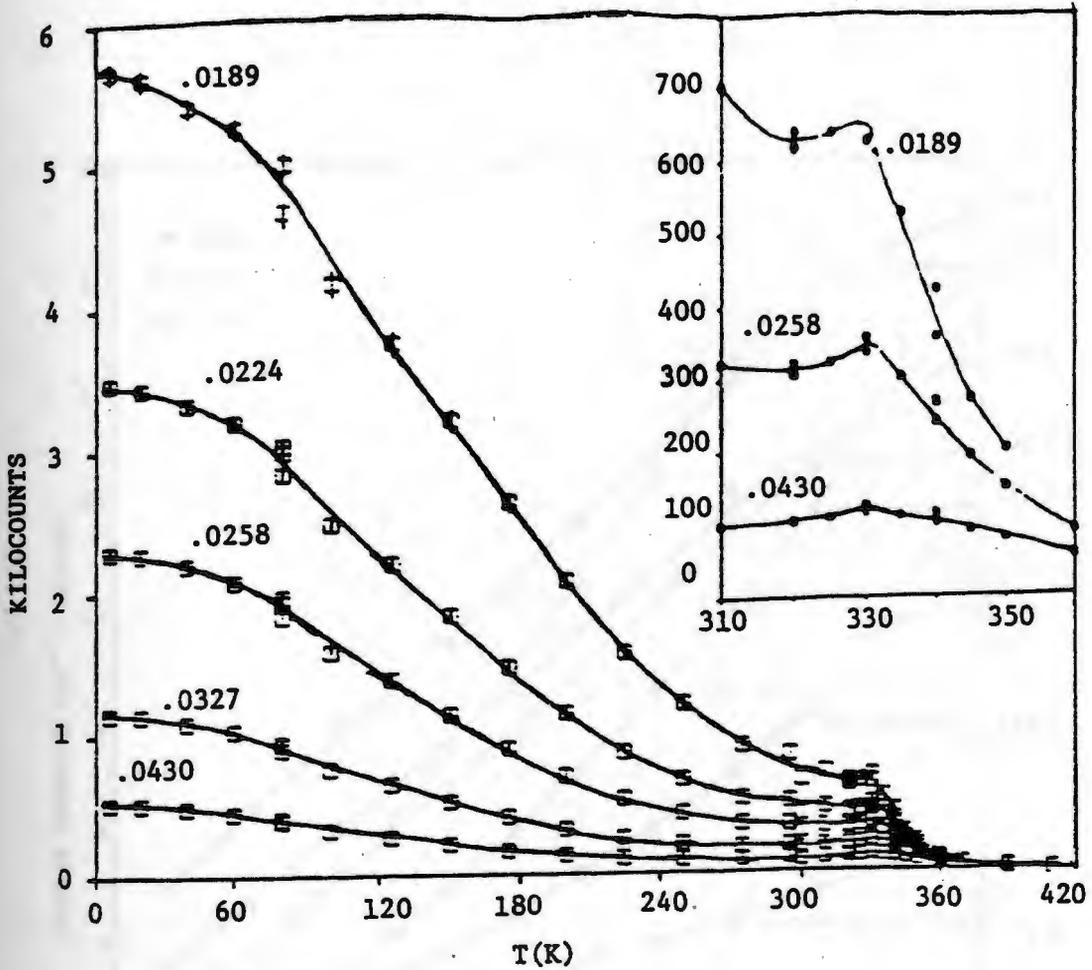


FIGURE 1

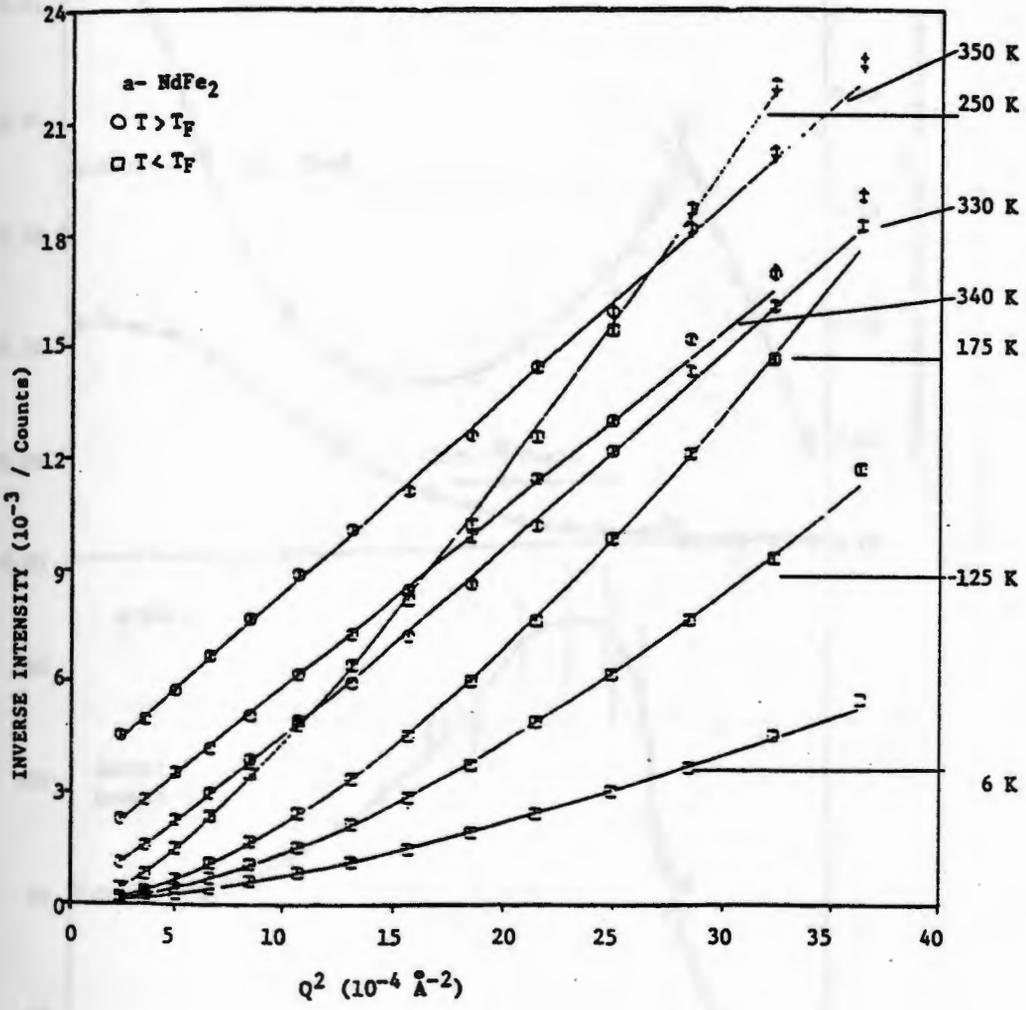


FIGURE 2

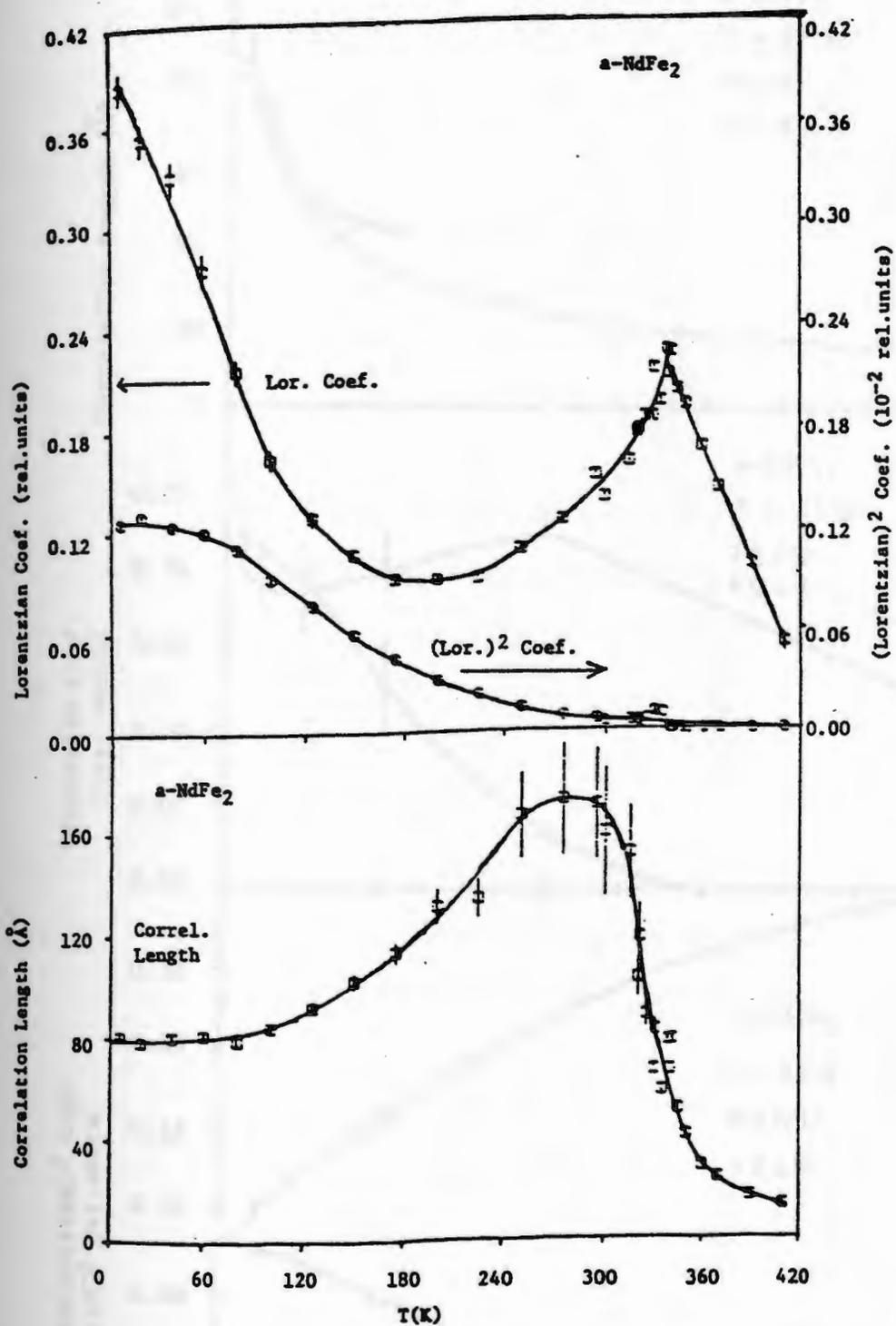


FIGURE 3

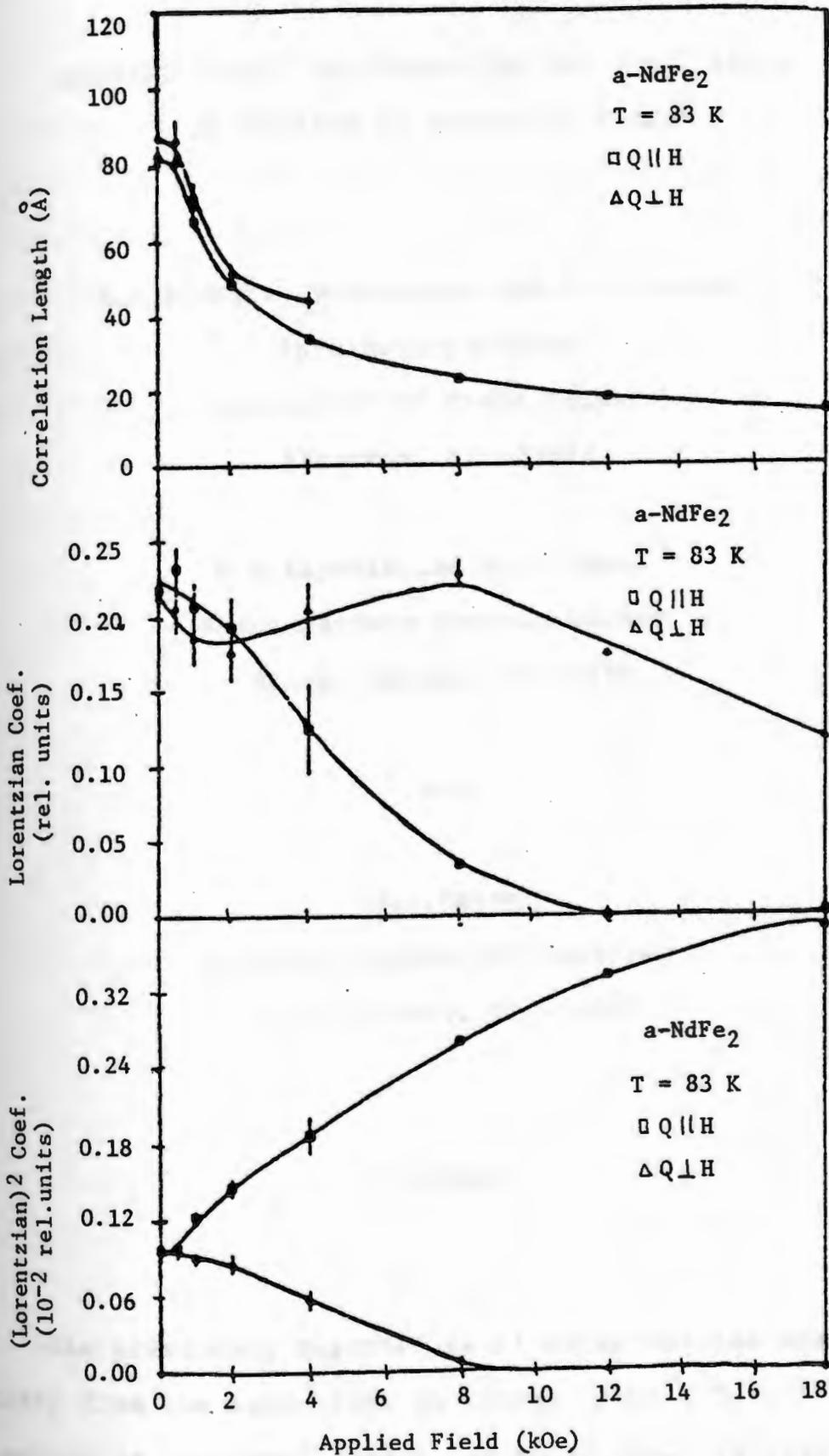


FIGURE 4

MAGNETIC FIELD DEPENDENCE OF THE SMALL ANGLE
SCATTERING IN AMORPHOUS DyCu

S.J. Pickart, S. Hasanain and D. Andrauskas

Physics Department

University of Rhode Island

Kingston, RI 02881

H.A. Alperin and M.L. Spano

Naval Surface Weapons Center

Silver Spring, MD 20910

and

J.J. Rhyne

National Bureau of Standards

Gaithersburg, MD 20899

ABSTRACT

The previously reported small angle neutron scattering (SANS) from the spin glass amorphous DyCu ($T_f = 17K$) was examined in applied fields up to 15 kOe. Application of

the field caused a decrease of the total SANS intensity and a decrease of the correlation length derived from the Lorentzian lineshapes to ~ 6 angstroms from the zero field value of 13.5 angstroms. Field-cooling and relaxation effects were also observed in the SANS intensity and are compared with similar phenomena in magnetization measurements.

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INTRODUCTION

Recently we reported [1] the observation of small angle neutron scattering (SANS) from the anisotropy-stabilized spin glass amorphous DyCu (a-DyCu), below its freezing temperature of 17K. Since magnetization measurements on this compound have revealed unusual field cooling and relaxation effects, we thought it would be interesting to explore the response of the SANS to an applied field, and also to note whether this response is different from other amorphous compositions [3-4] with stronger exchange forces.

EXPERIMENT

The measurements were performed at the NBS Reactor SANS facility using neutrons of 5.5 angstrom wavelength, which allows a useable range of scattering vectors $.015 \leq q \leq .105 \text{ \AA}^{-1}$. The sample, a sputtered film of 150 micron thickness, was placed in the tail of a variable temperature Dewar that could be inserted between the poles of an electromagnet with a maximum field of 19 kOe, applied in a horizontal direction in the plane of the sample. The majority of the measurements were made at 4K, with excursions to the neighborhood of 60K to determine background levels and investigate field cooling effects.

RESULTS AND DISCUSSION

The most obvious effect of the applied field was a sharp decrease of the total SANS intensity, as depicted in Figure 1, which shows data taken after cooling in zero field. Upon removal of the field and going through a demagnetization cycle, the intensity reverted to its initial value. Different behavior was observed when the sample was cooled through T in a field of 4 kOe. As indicated in the figure, the intensity resulting from this procedure was smaller, and, after removal of the field, did not regain its original value. However, after a time lapse of about 10 minutes the intensity had relaxed considerably towards its zero field value and eventually reached essentially the same level.

Accompanying this decrease in intensity was a marked decrease in the correlation length, as shown in Figure 2. These values were determined from an analysis of the lineshapes, which were Lorentzian in all cases to within the precision of the data, i.e. $I = A/(q^2 + K^2)$, where K is the inverse correlation length. What is plotted is the value of $(1/K)$ determined by least squares fitting of the above expression to the results of a circular average of the intensity over the 2-dimensional detector. The correlation length decreases to about 6 angstroms from its zero field value of 13.5 angstroms and then appears to make a slight but not necessarily statistically significant

rise.

Although these results by no means paint an unambiguous picture of the magnetization distributions in this compound, they suggest several qualitative features and mechanisms.

The decrease in scattered intensity observed here is similar to that observed in the amorphous compositions $TbFe_2$ reported by Rhyne and Glinka [3], and $NdFe_2$ reported by Spano et al [4]. Both these compositions are different in that, because of the high iron concentration, they display higher ordering temperatures and larger magnetic moments. In those cases the decrease in SANS intensity was tied to formation of an infinite percolating ferromagnetic cluster, which, since its lineshape is a delta function in q -space, removes intensity from the window seen by the detector. Undoubtedly, this is occurring in the present instance as well, since we know from magnetization measurements [2] that a large moment is developed by a field of a few kOe.

In the previous work on $TbFe_2$ [3] significant anisotropy was found in the scattering contours on application of the external magnetic field. The zero field circular constant intensity contours became elliptical in a field with the major axis along the field direction. Counting statistics on this very small sample of DyCu precluded a quantitative determination of any scattering

anisotropy, however within experimental error, no scattering anisotropy was observed.

Our final comment refers to the relaxation effects observed in the field cooled experiment, which we believe are the first example of such effects observed in SANS. Similar time dependent effects observed by Coey et.al.[2] in the remanent moment of this composition, but the time constant for the situation most closely related to the present experiment is much longer. Since the two experiments do not measure precisely the same quantity (the neutrons are sensitive to the q -dependent susceptibility while the magnetization measures $q = 0$) it is entirely possible that the two effects are driven by the same basic relaxation mechanism.

The work at the University of Rhode Island was supported by the National Science Foundation Grant Number DMR8207078. The work at NSWC was supported by the Office of Naval Research and by the NAVSWC Independent Research Fund.

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FIGURE CAPTIONS

FIGURE 5

Dependence of the circularly averaged total SANS detector counts (sample plus background) on applied field. The dashed line indicates the background level as obtained at 56K. Data were taken at 4K after cooling in zero applied field and in a 4 kOe field in the sequence indicated by the arrows. Time dependent relaxation effects were observed in the field cooled data after the field was removed as indicated.

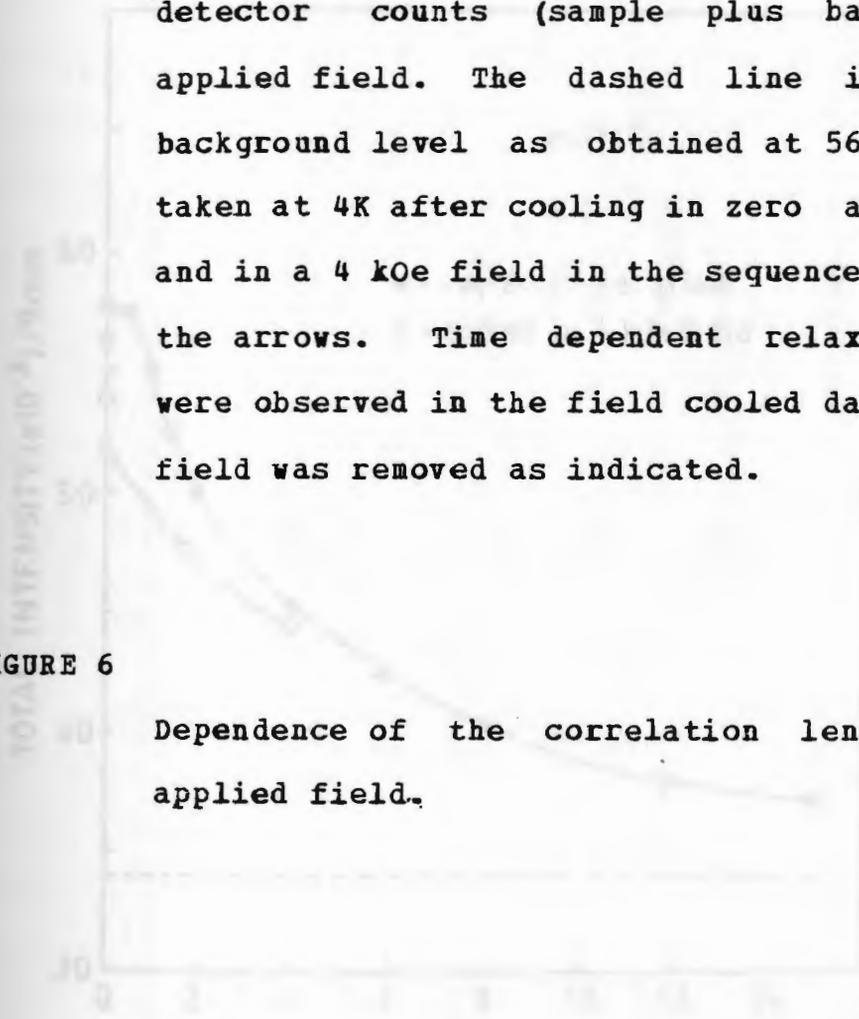


FIGURE 6

Dependence of the correlation length ($1/K$) on applied field.

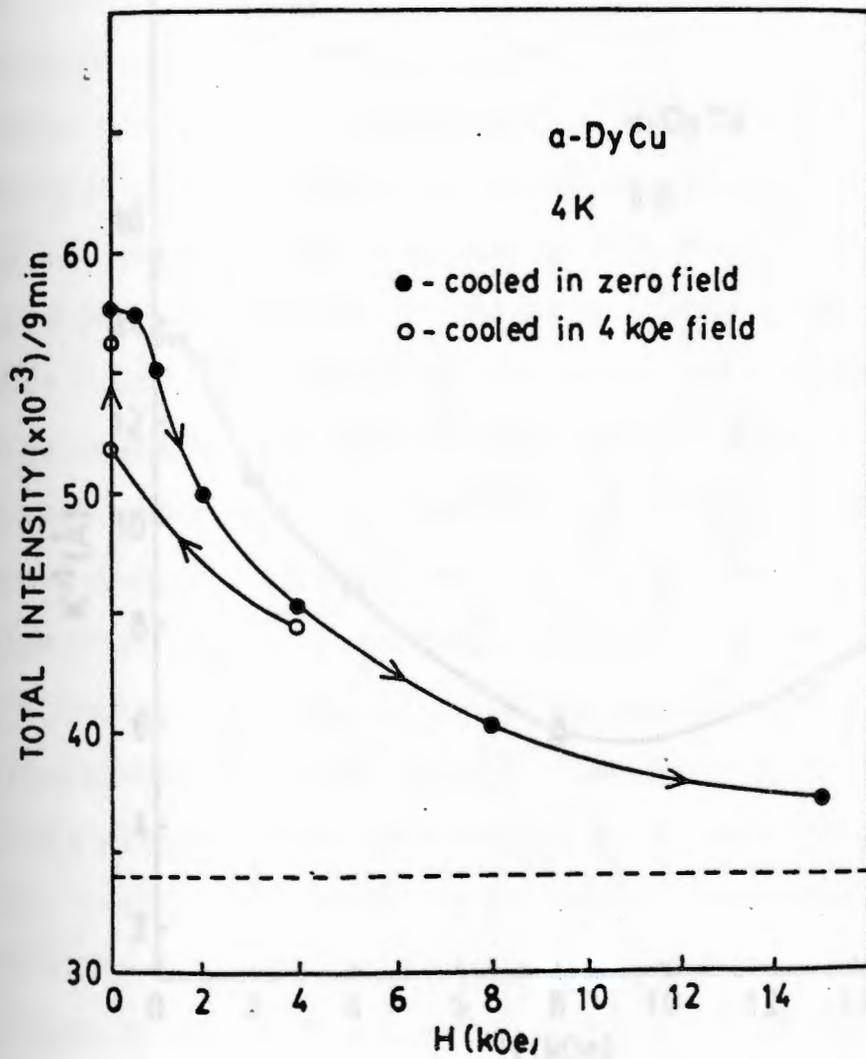


FIGURE 5

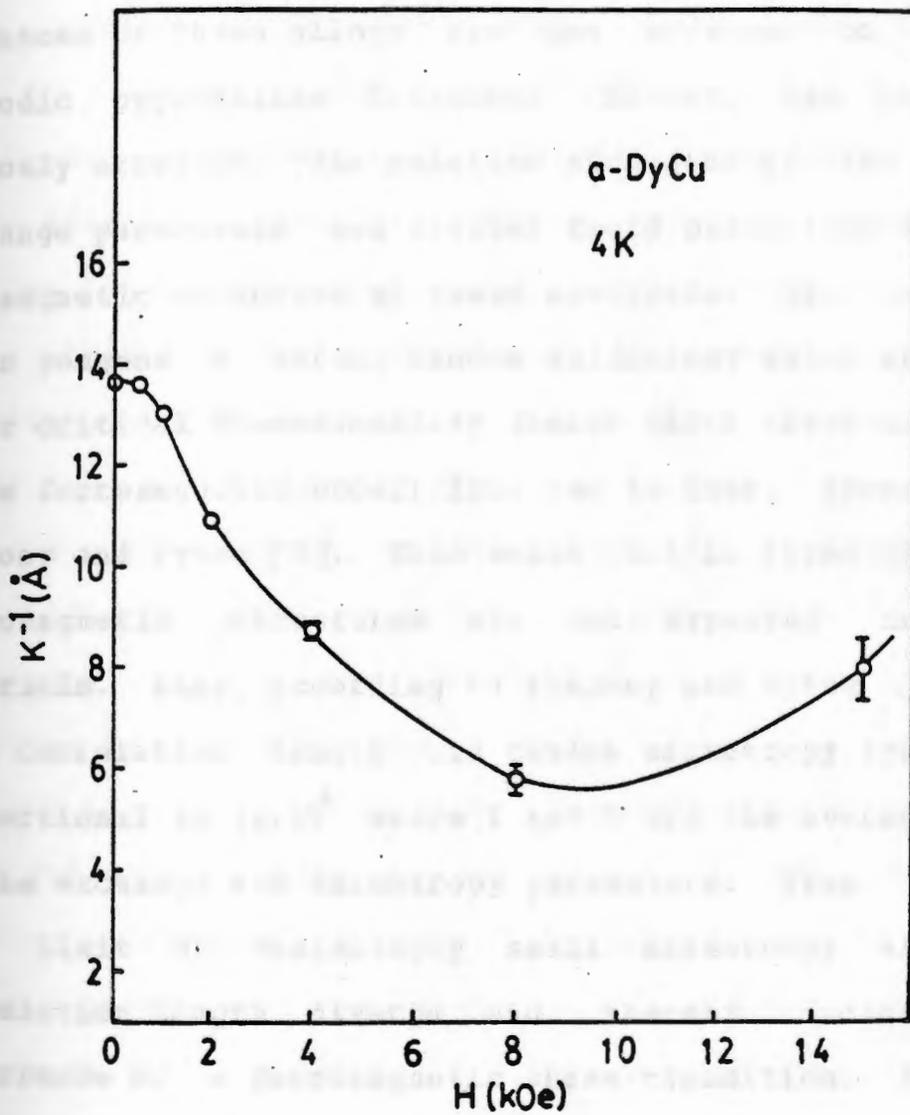


FIGURE 6

A. INTRODUCTION

Magnetism in amorphous rare-earth transition - metal alloys is influenced by exchange interactions and random magnetic anisotropy. Here, the term "amorphous" means that the atoms of these alloys are not arranged on regular, periodic crystalline lattices. Rather, the atoms are randomly arranged. The relative strengths of the various exchange parameters and crystal field parameters determine the magnetic structure of these materials. The rare-earth atoms possess a strong random anisotropy which shifts the lower critical dimensionality (below which there is no long range ferromagnetic order) from two to four, according to Aharony and Pytte [1]. This means that in three dimensions ferromagnetic structures are not expected in these materials. Also, according to Aharony and Pytte [1], the spin correlation length, in random anisotropy systems, is proportional to $(I/D)^2$ where I and D are the average values of the exchange and anisotropy parameters. Thus, only in the limit of vanishingly small anisotropy will the correlation length diverge and thereby indicate the occurrence of a ferromagnetic phase transition. In strong random anisotropy systems, the correlation length is expected to remain finite. In our materials, complex magnetic structures, discussed in the appendix on amorphous magnetism, are expected because of these effects.

Two amorphous rare-earth transition metal alloys that

are useful in studying the effects of random magnetic anisotropy on magnetic order are DyCu and NdFe₂. DyCu, an alloy in which anisotropy is considered dominant, undergoes a magnetic phase transition at approximately 17 K [2]. NdFe₂, a random anisotropy system with stronger exchange from Fe, has a magnetic phase transition at a temperature of approximately 330 K [3]. Both the exact nature of these phase transitions and the resultant magnetic structures are not yet fully understood and this inconclusiveness has led to a variety of both experimental and theoretical studies. Previous magnetic studies and the purpose of the current study will now be discussed.

In a magnetization study on amorphous DyCu by Coey et al [4], a sharp cusp in the low field magnetic susceptibility was seen indicating some sort of phase transition. Unusual field cooling and time relaxation effects in the magnetization measurements were also observed, for instance, the onset of low temperature irreversible susceptibility at the transition temperature. Saturation of the magnetization was not achieved even at fields on the order of 150 kOe, which indicates that the ordering was not ferromagnetic. These effects suggested that the alloy had random and anisotropic spin freezing which suggested an asperomagnetic spin glass-like magnetic structure below its transition temperature.

Additional evidence that some sort of phase transition

occurs in amorphous DyCu was found in a small-angle neutron scattering (SANS) experiment performed on this alloy by Pickart et al [2]. In this study, a break was observed in the zero field scattering patterns at a temperature roughly that of the transition temperature thus indicating some sort of phase transition.

Previous studies on the other amorphous alloy, NdFe₂, have also provided information on magnetic behavior in a random anisotropy system. As with DyCu, the magnetization curves for NdFe₂ displayed a lack of saturation even with fields on the order of 18 kOe [3]. Results from a magnetization study performed by Taylor, McGuire, Coey and Gangulee [5] led them to postulate for Nd-Fe alloys a sperimagnetic structure in which the spins of both the neodymium and the iron sublattices are non-collinear and the net moments of the sublattices are coupled in the same direction.

The current study was undertaken to obtain additional information about the nature of the phase transition in these materials and their resultant magnetic structures. To investigate the magnetic features that cause the observed field cooling and time relaxation effects in DyCu, the response of the SANS data to an applied field was recorded. The magnetic features of NdFe₂ were also investigated by SANS measurements as a function of applied field and temperature. The results are reported in this

thesis-SIGNIFICANCE OF SPONTANEOUS METALLIC GLASSES

1. Introduction

Silicate glass (ordinary window glass) does not crystallize when cooled from its liquid state to room temperature; it retains the disordered structure of a liquid, while a metal crystallizes rapidly when cooled from its molten state. In 1938, Dorem and coworkers at the California Institute of Technology succeeded in cooling a molten alloy of gold and silver sufficiently rapidly to prevent crystallization and to retain it at room temperature [1]. When they showed that this alloy had the disordered structure of a liquid and was a good conductor of electricity, a property associated with metals, a new class of materials - metallic glasses - came into existence.

Metallic glasses, also called amorphous metallic alloys, glassy alloys, or non-crystalline alloys are materials that do not have the long range spatially periodic arrangements that crystalline materials have but they have short range atomic order. Since the state of lowest energy is usually an ordered state, these alloys are generally metastable with respect to some crystalline phase or phases and spontaneous recrystallization is inhibited by nucleation barriers which slow down the formation of stable crystalline nuclei and/or by kinetic barriers which limit the rate at which existing stable crystalline nuclei can

B. SIGNIFICANCE OF AMORPHOUS METALLIC ALLOYS

1. Introduction

silicate glass (ordinary window glass) does not crystallize when cooled from its liquid state to near room temperature; it retains the disordered structure of a liquid, while a metal crystallizes rapidly when cooled from its molten state. In 1959, Duwez and coworkers at the California Institute of Technology succeeded in cooling a molten alloy of gold and silicon sufficiently rapidly to prevent crystallization and in retaining it near room temperature [6]. When they showed that this alloy had the disordered structure of a liquid but was a good conductor of electricity, a property associated with metals, a new class of materials - metallic glasses- came into existence.

Metallic glasses, also called amorphous metallic alloys, glassy alloys, or non-crystalline alloys are solids that do not have the long range spatially periodic atomic arrangements that crystalline materials have but may have short range atomic order. Since the state of lowest free energy is usually an ordered state, these alloys are generally metastable with respect to some crystalline phase or phases and spontaneous recrystallization is inhibited by nucleation barriers which hinder the formation of stable crystalline nuclei and/or by kinetic barriers which limit the rate at which existing stable crystalline nuclei can

grow [7,8]. These processes are temperature dependent and place an upper limit on the temperature at which metallic glasses can be used.

Theories of magnetic structure that are based on crystalline lattices cannot be satisfactorily applied to the structure of amorphous metallic alloys. Their predictions do not agree well with experimentally obtained structural data, as is to be expected since this data (usually obtained by X-ray, electron, or neutron scattering) yield information only about short range order. This lack of agreement has led scientists to construct models to describe the structure of these alloys. One such model is the DRPHS (dense random packing of hard spheres) model developed by Bennett and others [9]. Here, computer construction methods are used to develop a model that is consistent with packing constraints.

Knowledge about the structure of amorphous metallic alloys is important to both scientists and engineers. Engineers are interested in the structure of these materials because of the relationship between properties and the structure, scientists because this knowledge will lead to a better understanding of the solid state.

Some classes of metallic glasses [10] are (1) the transition metal - metalloid (TM-M) alloys, (2) the transition metal - zirconium or hafnium (TM-Zr-Hf) alloys, (3) the simple metal alloys, such as magnesium-zinc and

calcium-magnesium and (4) the rare earth-transition metal (RE-TM) alloys. Typically, the TM-M alloys are composed of iron, nickel, or cobalt combined with a metalloid such as boron, carbon, silicon, phosphorus, or aluminum. The metalloid, which usually lies in the 15-30 atomic percent range, is necessary for the formation of the amorphous phase since it lowers the melting point so that the alloy can be quenched rapidly enough to form the amorphous phase. Usually, the TM-Zr-Hf alloys are composed of iron, nickel, or cobalt combined with about 10 atomic percent of zirconium or hafnium. Iron is often used as the transition metal in both these types of alloys since it is inexpensive and it usually provides useful magnetic properties, such as high permeability.

The RE-TM alloys are composed of a rare earth such as dysprosium, neodymium, terbium, or gadolinium and a transition metal such as iron, copper, or cobalt. These alloys can be produced in amorphous form over a wide range of compositions which makes them attractive to scientists since it allows them to study variations in structure and properties over a wide range of compositions of the same alloy without having to worry about compound formation. Also, these metallic glasses are particularly interesting because complex magnetic orderings are expected in certain compositions of these materials because of effects such as random anisotropy.

2. Preparation

Preparation techniques for the formation of amorphous metallic alloys can be grouped into two categories: (1) direct quenching from the melt (process of solidification by rapid cooling from the liquid state) and (2) deposition techniques (techniques in which the solid is not obtained directly from the liquid state). Deposition techniques include electrolytic deposition, vacuum deposition and rf (radiofrequency) sputtering, a technique in which the atoms of a material pass directly from vapor to a solid on a cold substrate. Direct quenching from the melt involves such techniques as "splat cooling" - a process in which a molten drop is shot onto a cold substrate where it forms a splat or thin film and "jet-casting" or "melt-spinning", a technique used in the formation of a continuous ribbon of metallic glass [11]. These techniques involve cooling rates of 10^6 degrees C per second or higher which are necessary in order to avoid crystallization. Many metallic glasses for technological application are formed by melt-spinning.

3. Applications

Many metallic glasses have high strengths and good ductibility. The combination of these properties has led to the use as brazing filler materials. Ribbons of these

materials are used to join together joints in aircraft engines as well as other devices. Another use that is being considered is as reinforcement for concrete or other building materials. Also, their high degree of hardness makes these alloys good candidates for cutting edges on tools.

Some metallic glasses can become superconducting at low temperatures, which, in combination with their ductibility, would make them useful in superconducting wires.

Some iron-chromium glasses form a protective film on their surfaces when exposed to aqueous electrolytes. These glasses are then resistant to chlorides (such as sea water) and sulfates which makes them attractive for use in marine and medical environments as cables, chemical filters, and scalpels among other things.

One area of large application for these alloys is as transformer cores. The properties of low power losses and low coercivity as well as the ease of large scale production by melt-spinning make them attractive for this application. Also, high permeability and high yield stress has led to the utilization of these alloys as magnetic shielding.

In particular, the amorphous RE-TM alloys are being considered for use as magnetic bubbles for the storage of data in computer memory because of their low thermal

conductivity and lack of grain boundaries, which are requirements for the storage of data. By using these alloys, the size of the data storage area can be reduced thereby leading to smaller components for computer memory devices, an ongoing concern of many engineers. The large spin-disorder scattering also makes them attractive as magneto-resistance sensors.

C. PHASE TRANSITIONS AND AMORPHOUS MAGNETISM

1. Phase Transitions

When a system changes from one phase or state to a different phase or state at a well-defined temperature it is said to undergo a phase transition. This occurs because of a competition between ordering forces (such as cohesion forces) and disordering forces (such as thermal fluctuations). For example, in a magnetic system, when the ordering forces begin to dominate, the system undergoes a change from a high temperature magnetically disordered phase to a low temperature magnetically ordered phase at the transition temperature. Also, in a fluid system, such as water, the transition from the liquid to the solid state is a phase transition that occurs when the ordering forces begin to dominate.

2. Amorphous Magnetism

Conventional theories of magnetism involve solids that have their magnetic atoms arranged on regular crystalline lattices. Because of this periodic arrangement, the resultant crystalline magnetic orderings in these solids contain both long-range spatial order as well as long-range magnetic order- this results in all atomic sites being equivalent in these materials as well as all macroscopic directions being equivalent in the bulk. These orderings

are described by the following types of magnetism - ferromagnetism, antiferromagnetism, and ferrimagnetism. The disordered phase of these materials, usually the high temperature phase, is described by paramagnetism. Much has been written about these topics. However, these results do not provide a good description of the magnetic behavior of amorphous materials.

Magnetism in amorphous materials cannot be adequately described by conventional theories because the random arrangement of their atoms leads to new magnetic structures. These new magnetic structures lack long-range spatial order which results in no atomic sites being equivalent but all macroscopic directions being equivalent in the bulk which is fundamentally different from the basis of conventional theories.

Magnetism in amorphous materials is influenced by exchange interactions between neighboring atoms and randomly distributed local magnetic anisotropies. Exchange interactions can be either direct or indirect. Direct exchange is a coupling between atoms that have overlapping wave functions - it is strong but short-range. Indirect exchange coupling acts between atoms that are far enough apart to require an intermediary. In metals, conduction electrons are the intermediary and the coupling is known as the RKKY interaction. In insulators, nonmagnetic ions are the intermediary and the coupling is known as

superexchange. The randomly distributed local magnetic anisotropies are due to crystal field effects which will be detailed later.

The theory of magnetism of these materials examines the energy of these magnetic interactions. Using the assumption that the exchange interaction is isotropic and dependent only on the distance between interacting atoms, the exchange Hamiltonian can be written as

$$H = - \sum_{i,j} J(r_{ij}) \vec{S}_i \cdot \vec{S}_j$$

where r is the distance between atoms i and j , \vec{S}_i and \vec{S}_j are the spins of atoms i and j , and J is the exchange parameter [12]. Since the only structural information required by this approximation is an interatomic distance, it works well when applied to amorphous metals in which the exchange has no angular or directional dependence. Consequently, this approximation is not good when applied to amorphous insulators in which the exchange does have angular or directional dependence. Also, the above Hamiltonian does not contain any information about crystal field effects.

Crystal field effects arise from the existence of an anisotropic electric field at each atomic site. Because this electric field varies from point to point in an amorphous system, each magnetic atom will have a preferred

direction in which to align its spin given by the direction of the local free energy minimum. Using the assumption that there is one local easy axis at each site (case of uniaxial symmetry), a Hamiltonian that takes into account both magnetic anisotropies and exchange interactions can be written as

$$H = - \sum_i \vec{D}_i (S_z)_i^2 - \sum_{i,j} J(r_{ij}) \vec{S}_i \cdot \vec{S}_j$$

where the second term has been previously described, \vec{D} is the axial crystal field strength, and S_z is the total spin of the atoms along the local easy direction z [12]. The orientation of individual spins is determined by competition between the exchange and anisotropy interactions and competition among the exchange interactions [13]. Then, different relative strengths of J and D in the above Hamiltonian result in different magnetic structures for these materials.

The resultant magnetic structures for these materials are ferromagnetic, antiferromagnetic, speromagnetic, and asperomagnetic. Both these structures and the above discussion involve materials that have one distinct magnetic sublattice. Here, the term "sublattice" refers to a group of atoms that have the same magnetic interaction and are the same magnetic element. One such group of

materials are the transition metal-metalloid alloys. Descriptions of the magnetic structures listed above will now follow.

Amorphous ferromagnetism results when the exchange dominates the anisotropy ($|\vec{J}| \gg |\vec{D}|$) and the exchange parameter is positive ($J > 0$). In an amorphous ferromagnet, the spins align along the same direction below the transition temperature. This alignment results in a spontaneous magnetization which decreases with increasing temperature and, in the absence of an applied magnetic field, disappears at the transition temperature. Above the transition temperature, the spins are no longer aligned and the material becomes a paramagnet. Some examples of amorphous ferromagnets include transition metal-metalloid alloys of varying compositions of Fe-B, and (Fe-Ni)-B-P [12].

Amorphous antiferromagnetism results when the exchange dominates the anisotropy ($|\vec{J}| \gg |\vec{D}|$) and the exchange parameter is negative ($J < 0$). Because the exchange parameter is negative, when the spin of one atom is aligned in a given direction, the neighboring spins will want to align themselves in an exact opposite direction. This antiparallel alignment of spins means that there is no spontaneous magnetization present in these materials in the absence of an applied magnetic field. An applied magnetic field induces some degree of magnetization in these

materials. Above their transition or Neel temperature, thermal effects exceed the exchange and these materials also become paramagnets. One amorphous antiferromagnet that has been reported is a high iron content glass [14].

Amorphous speromagnets resemble random antiferromagnets. In these materials, the spins of the magnetic sublattice are frozen into random orientations below the transition temperature. This structure has short-range correlations of spin directions on the order of the nearest neighbor distances but no long-range correlations. There is no spontaneous magnetization present in these materials due to these factors. Speromagnetism is distinguishable from paramagnetism, the magnetic structure of these materials above their transition or spin-freezing temperature, since, in paramagnets, the directions of the spins fluctuate constantly and randomly with time while, in speromagnets, the directions of the spins are frozen into orientations that do not vary with time [12].

Asperomagnets can be thought of as random ferromagnets. In this magnetic structure, the spins are also frozen in random orientations that do not vary with time. However, some orientation(s) are more likely to occur than others which gives rise to a spontaneous magnetization which disappears at the transition temperature. Above this temperature, these materials

exhibit paramagnetic behavior. One amorphous alloy that has shown asperomagnetic behavior is DyNi_3 [13].

Amorphous magnetic materials that have two distinct magnetic sublattices, such as the rare-earth transition-metal alloys, are also of interest. Using the same assumptions as before, the Hamiltonian for these alloys can be written as

$$H = -\sum_a \vec{D} (S_3)_a^2 - \sum_b \vec{D} (S_3)_b^2 - \sum_{aa'} J_{aa'} \vec{S}_a \cdot \vec{S}_{a'} - \sum_{bb'} J_{bb'} \vec{S}_b \cdot \vec{S}_{b'} - \sum_{ab} J_{ab} \vec{S}_a \cdot \vec{S}_b$$

where a and b are the indexes of sites on the rare-earth transition-metal sublattices respectively, J_{ab} is the exchange parameter for the interaction between the rare-earth and the transition-metal atoms, and the other parameters are the same as before [15]. As before, the different relative strengths of the exchange parameters and the crystal field strengths result in different magnetic structures. These resultant magnetic structures include ferrimagnetic and sperimagnetic.

In amorphous ferrimagnets, each distinct magnetic sublattice has a collinear alignment of spins and these alignments point in opposite directions. Since the net moments of these two sublattices are unequal, there is a resultant spontaneous magnetization. Below the transition

temperature, the temperature dependence of this magnetization is qualitatively similar to that observed in ferromagnets. Quantitatively, this magnetization decreases more rapidly with increasing temperature. Above the transition temperature, these materials also exhibit paramagnetic behavior. One typical ferrimagnet is thought to be amorphous GdCo_3 [13]. In this alloy, there is strong positive Co-Co exchange coupling which produces a ferromagnetic cobalt sublattice. The Co-Gd coupling is negative and much weaker giving rise to a gadolinium sublattice that is coupled antiparallel to the cobalt. Thus, the resulting structure for this alloy is ferrimagnetic.

Amorphous sperimagnets can be regarded as canted or noncollinear ferrimagnets. In these materials, one of the magnetic sublattices usually has its spins aligned parallel as in a ferromagnet while the other magnetic sublattice has its spins frozen into random orientations that are not collinear. There are two types of sperimagnets that are distinguishable. For illustrative purposes, the rare-earth transition-metal alloys, DyCo_3 and NdCo_3 , will be examined. In the one type, DyCo_3 [15], it has been proposed that the spins of the cobalt sublattice are oriented parallel while the spins of the dysprosium sublattice are oriented randomly in a sphere. This results in the net moments of the two sublattices being antiparallel with the dysprosium

moments oriented randomly within a cone. In the other type, NdCo_3 , the spins of the cobalt sublattice are thought to be parallel while the neodymium spins are oriented randomly in a hemisphere. This results in a parallel alignment of the net moments of the two sublattices with the neodymium moments oriented randomly within a cone.

Two other magnetic structures that also have been suggested for amorphous metallic alloys are the superparamagnetic state and the spin glass state.

In a sample of magnetic material, as the applied magnetic field strength increases, the size of the three-dimensional correlated networks of nearest neighbor moments ("clusters") may increase until an infinite cluster of correlated spins forms in the sample. The point at which this infinite cluster begins to form is indicated experimentally when the value of the spin correlation length becomes infinite and thus divergent. As the field is further increased beyond this point, the number of magnetic atoms belonging to this infinite cluster may also increase. However, some magnetic atoms may still belong to smaller clusters whose magnetic moments within the cluster are correlated but whose net cluster moment is free to fluctuate in many directions. The result is superparamagnetic behavior, which differs from paramagnetic behavior in that the effective moment of a cluster results from the sum of the moments of all the magnetic atoms

within a cluster while a moment in a paramagnet arises from an individual magnetic atom. Two experimental features characterize superparamagnetism. One feature is that there is no hysteresis in the field dependence of the magnetization and the other is that the magnetization is a universal function of H/T [12].

The spin glass state is one in which no long-range magnetic order occurs. Rather, the spins of the magnetic atoms are frozen into random orientations that do not fluctuate with time. Short-range correlations between spins cause the formation of spin clusters in this state and this spin glass behavior arises from a competition between the random distribution in the magnitude and the sign of the interactions between the neighboring spins [12]. Some characteristic spin glass alloys are the crystalline alloys of type Cu-Mn. One amorphous magnetic alloy that is considered a concentrated spin glass is YFe [16].

Some experimental observations that are typical of spin glass behavior are

- (1) a cusp in both the ac and dc susceptibility at the transition or spin-freezing temperature [12]
- (2) sensitivity of the susceptibility to applied magnetic field at and near the transition temperature. The application of a field on the

order of a few tens of gauss completely smears out the susceptibility cusp. Also, time dependent and history dependent effects are observed in the low-field magnetization [17].

(3) the absence of an anomaly, which is expected for a phase transition, in the specific heat. The extra specific heat due to the impurities is a linear function of T , $C = \gamma T$, where γ is independent of concentration and much larger than the ordinary term of normal metals [18].

(4) for low concentrations of the magnetic element (the "impurity"), quantities such as the magnetization, saturation remanence, and molar heat capacity obey scaling laws and are universal functions of the reduced variables H/c and T/c . For instance, $M/c = f(H/c, T/c)$ where c denotes the concentration of the magnetic element [17].

(5) In SANS studies, the spin correlation length remains finite at and below the spin-freezing temperature [16].

The theories of amorphous magnetism described above are models that are based on fitting magnetization data to theoretical predictions. Neutron diffraction and SANS experiments are performed to obtain additional microscopic structural information about these alloys. This information is then compared to the models to check the

validity of these models.

1. Neutron Scattering

In this section, scattering theory will be reviewed briefly and experimental aspects of both neutron scattering and small-angle neutron scattering will be discussed, emphasizing their applicability to the study of the magnetic behavior of materials.

The technique of neutron scattering has been used in many studies of the static and dynamic properties of materials. The usefulness of this technique arises from both the properties of the neutron and the interaction of the neutron with the target sample or scatterer. Some of the properties of the neutron are: (a) the wavelength corresponds to interatomic distances, (b) the energy of the neutron is frequently on the order of the energy of excitations in condensed matter, (c) the neutron is a neutral particle, and (d) the neutron has a magnetic moment. The interaction of the neutron with the sample consists of two separate parts [10]. First, there is the neutron-nucleus force, which is a very strong and very short-range force that is responsible for the binding of neutrons and protons in nuclei. Second, the magnetic dipole-dipole interaction of the neutron with the magnetic moments of the atoms in the sample is responsible for the magnetic scattering.

D. PROCEDURES AND METHODOLOGY

1. Neutron Scattering

In this section, scattering theory will be outlined briefly and experimental aspects of both neutron scattering and small-angle neutron scattering (SANS) will be discussed, emphasizing their applicability to the study of the magnetic behavior of materials.

The technique of neutron scattering has been used in many studies of the static and dynamic properties of materials. The usefulness of this technique arises from both the properties of the neutron and the interaction of the neutron with the target sample or scatterer. Some of the properties of the neutron are (a) the wavelength corresponds to interatomic distances, (b) the energy of the neutron is frequently on the order of the energy of excitations in condensed matter, (c) the neutron is a neutral particle, and (d) the neutron has a magnetic moment. The interaction of the neutron with the target sample consists of two separate parts [19]. First, there is the neutron-nuclear force, which is a very strong and very short-range force that is responsible for the binding of neutrons and protons in nuclei. Second, the magnetic moment of the neutron interacts, by ordinary dipole magnetic forces, with the magnetic moment of the electrons

in the target sample .

As a result of properties (a) and (b), neutron scattering provides a sensitive means of studying the energy spectra of elementary excitations in materials. Because of (c), the absorption of neutrons by matter is often very small, and large sample volumes can be studied by this technique [20].

Neutron scattering will be sensitive to the arrangement of nuclei in a sample and will thus provide information about the structure of the sample because of the nuclear interaction. The magnetic interaction makes this technique sensitive to the magnetic properties, so that neutron scattering can be used to study the magnetic structure of a material.

The scattering of neutrons is characterized by the change of energy $\hbar\omega$ and momentum $\hbar\vec{Q}$. \vec{Q} , which is the scattering vector, is defined as $\vec{Q} = \vec{k}_0 - \vec{k}_f$ where \vec{k}_0 and \vec{k}_f are the incident and final neutron wave vectors respectively. This relation holds for both elastic and inelastic scattering, but for elastic scattering, the scattered energy is the same as the incident energy, $|\vec{k}_0| = |\vec{k}_f|$, and $Q = (4\pi \sin\theta) / \lambda$ where λ is the incident neutron wavelength and 2θ is the scattering angle.

Neutron elastic scattering diffraction techniques yield scattering patterns in which the scattered intensity, I , is determined as a function of the scattering vector, Q .

This can be accomplished in two ways (1) Variable 2θ -Method -- Measurement of the diffraction angle by using a constant neutron wavelength. and (2) Variable λ -Method -- Measurement of the scattered energy by using variable neutron wavelengths at constant scattering angle.

In this study, the technique of SANS, a variable 2θ -Method, was used to obtain the data. This technique was used because neutron scattering techniques that are used for investigating magnetic correlations in amorphous alloys require measurements at very small Q near the forward beam direction ($Q=0$) because this is the only point of in-phase scattering for an amorphous system [16].

The resultant SANS patterns contain contributions from nuclear and magnetic scattering. In order to investigate the magnetic features of the samples, the scattering of magnetic origin is separated out. Two experimental methods that can be used to separate the magnetic scattering are the following. One method is to subtract the scattering pattern taken at a temperature well above the transition temperature where the sample is in its disordered magnetic state from a scattering pattern taken well below the transition temperature. The resultant pattern will contain the magnetic contribution. This method was used in our study. Another method to extract the magnetic contribution is to make difference measurements with a magnetic field applied successively parallel and perpendicular to the

scattering vector and to keep the sample well below the transition temperature [21]. This method is used for samples that crystallize upon heating above their transition temperature, but is not feasible here because of the high fields required for saturation.

After the subtraction procedure, information about the magnetic features of a material can then be obtained from the resultant scattering pattern. For SANS data, a peak or break in the lineshape of the Intensity versus Temperature scattering patterns for different Q values indicates the occurrence of a phase transition. The shape of the Intensity versus Q curves also provides magnetic information. To obtain this additional information, curves are fitted to the experimental data both above and below the transition temperature. A fitted curve that has the Lorentzian form,

$$I(Q) = A/(Q^2 + K^2)$$

indicates that the scattering can be interpreted in terms of an inverse correlation length $K(T)$, the decay constant of a real space correlation function of the Ornstein-Zernicke form $((1/r)\exp(-Kr))$ [2]. Knowledge of this length provides information about the magnetic

ordering that is occurring in the sample.

2. Experimental Procedures

In this study, the data was obtained with a 64 X 64 squared centimeter area detector at the NBS Reactor using an incident neutron wavelength of 5.5 angstroms. To obtain the data, the amorphous DyCu sample was placed in the tail of a Dewar vessel in which the sample temperature could be varied. The tail of this vessel was placed between the poles of an electromagnet which was used to apply a magnetic field in a horizontal direction in the plane of the sample. The sample, which was now in the path of the neutron beam, was cooled down from room temperature to 4 K in zero field. While at 4 K, a magnetic field was applied and measurements at different field strengths were recorded. After the field was removed and the sample taken through a demagnetization cycle, the sample was heated to a temperature well above 17 K and a field was applied. In the presence of this field, the sample was again cooled down to 4 K and SANS data obtained. Next, the field was removed and data was again recorded to study the field cooled effects. After a suitable time interval, SANS data were again obtained to study the time relaxation effects. Measurements for the background were performed at about 60 K after the above process.

The NdFe_2 sample was placed in the neutron beam path in the same manner as the DyCu sample. The sample was cooled in zero field to a temperature below its transition temperature and SANS data were taken in zero field from 6 to 410 K. The sample was again cooled down in zero field to 83 K and measurements from 0 Oe to 18 kOe were taken at this temperature. As for DyCu, measurements for the background were performed at a temperature well above the transition temperature after this process.

E. RESULTS AND DISCUSSION

For amorphous DyCu, the dependence of the total SANS intensity on applied field shows a decrease in the total intensity with increasing field. Also, time dependent relaxation effects in the field-cooled data are shown on this curve. The spin correlation length vs. applied field shows a marked decrease in the correlation length with increasing field. The correlation length was obtained from the lineshapes which were Lorentzian at all temperatures to within experimental error.

For amorphous NdFe₂, the scattered neutron intensity vs. temperature curves for several values show a small peak at the transition temperature with an increase in the scattering at lower temperatures. Plots of the inverse intensity versus Q curves both above and below the transition temperature have Lorentzian lineshapes above the transition temperature and increasingly deviated from this form below this temperature. For this alloy, the temperature dependence of the spin correlation length has a broad maximum near to, but lower than, the transition temperature and shows a decrease in correlation length below this temperature. In addition, the SANS data obtained with an applied field shows a sharp decrease in intensity with an increasing field at constant temperature.

Curvature of the inverse intensity versus Q^2 curve indicates a departure from the Lorentzian form of the

scattering. Such departure is evidence of the absence of long-range ferromagnetic order. Another indication that ferromagnetic order is not present is the increase in scattering at low temperatures. If long-range ferromagnetic order were present, the SANS intensity would nearly vanish below the transition temperature since the scattering will occur only infinitesimally close to $Q = 0$ [22].

The theory of Aharony and Pytte [1] as applied by Spano and Rhyne [22] shows that the presence of the random anisotropy field makes it energetically favorable for such a system to form small regions of clustered spins thus inhibiting the transition to long-range order. In neutron scattering measurements, the effect of this formation is to convert the delta function scattering at $Q = 0$, a characteristic of long-range ferromagnetic order, to the sum of a Lorentzian plus Lorentzian squared. This sum has the form

$$I(Q) = F/(Q^2 + K^2) + G/(Q^2 + K^2)^2$$

where $K(T)$, the inverse correlation length, is nonzero below the transition temperature because the correlation length remains finite in strong random anisotropy systems.

For amorphous NdFe_2 , the lineshapes of the inverse intensity versus Q curves below the transition temperature have the Lorentzian plus Lorentzian squared form. This data suggests that the presence of the random anisotropy prevents the spins from correlating beyond a certain length. Thus, an infinite cluster of spins, a characteristic of ferromagnets, does not form but rather short-range correlation results in the formation of clusters of spins. Since such an arrangement of spins is one feature of the spin glass state, amorphous NdFe may be considered spin glass-like.

In addition, for amorphous NdFe_2 , the applied field dependence is also different from that expected for a ferromagnet. The behavior of this alloy is similar to that exhibited by a system of super-paramagnetic spins or clusters. Thus, an examination of the field dependent data result in this alloy being considered a system of spin clusters of varying size, whose larger clusters are driven into a near-infinite ferromagnetic cluster by the application of an external field. Since this cluster will scatter only infinitesimally near to $Q = 0$, only the smaller clusters will contribute to the SANS patterns. This accounts for the resultant decrease in scattering with the application of a field.

The applied field dependence that is seen in amorphous DyCu is similar to that observed in TbFe_2 [23] and NdFe_2

[24]- In these alloys, this behavior was attributed to the formation of a near-infinite ferromagnetic cluster with regions of smaller clusters that do not become part of the near-infinite cluster. The relaxation effects observed after the field-cooling in DyCu are believed to be the first observance of such effects in SANS.

F. SUMMARY

The SANS measurements demonstrate that amorphous NdFe₂ and DyCu do not have a magnetic phase transition to long-range ferromagnetism. They indicate that these alloys undergo a transition to a state that has some features of a spin glass. This behavior is consistent with what is predicted for a strong random anisotropy system.

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