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MASTER**NEUTRON SCATTERING EXPERIMENTS ON THE MAGNETISM IN****Cu-Mn SINGLE CRYSTALS****S. A. Werner and J. W. Cable**

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Neutron scattering experiments on the magnetism in Cu-Mn single crystals^(a)

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ABSTRACT

We have carried out neutron polarization analysis experiments, and unpolarized neutron scattering experiments, on annealed single crystal Cu-Mn samples of 5, 10, 15 and 25 atomic percent Mn. Peaks in the magnetic diffuse scattering are found at the $[1, \frac{1}{2} \pm \delta, 0]$ -type positions in reciprocal space, where $\delta = 0.34, 0.29, 0.25$ and 0.19 for the 5, 10, 15 and 25 percent Mn samples respectively. This magnetic scattering is symmetry-related to the diffuse nuclear scattering centered at the $[1, \frac{1}{2}, 0]$ -type positions arising from atomic short range order. The intensity of this diffuse magnetic scattering in all of the alloys decreases smoothly with temperature, approaching zero in the range 300-350 K. The spin-spin correlation range, as judged from the width of the magnetic diffuse peaks, also decreases smoothly with temperature. Neither the width nor the intensity of this magnetic scattering gives any hint of a freezing transition at lower temperatures.

INTRODUCTION

The curious magnetic properties of Cu-Mn alloys in the α -solid solution range have attracted considerable attention for a very long time (1,2,3). In spite of the fact that a large number of neutron scattering experiments, (4,5,6,7,8) of increasing sophistication, have been carried out on these alloys, a full understanding of the static and dynamic magnetic structure of these materials has not yet evolved. Fundamentally, we are faced with the problem of understanding the origin of the cusp in the low-field magnetic susceptibility (9) occurring at some temperature dependent upon Mn concentration, in the absence of the development of long-range antiferromagnetic order at low temperatures. Since there is no abrupt change in the entropy at the temperature at which the susceptibility cusp occurs, now commonly called the freezing temperature T_f , the low temperature magnetic structure of these alloys is

thought to resemble the static (metastable) structure of a glass. This spin glass or mictomagnetic behavior now appears to be a general characteristic of many more or less disordered alloys [such as $\text{Au}(\text{Fe})$, $\text{Ag}(\text{Mn})$, $(\text{La}_{1-x}\text{Gd}_x)\text{Al}_2$], irrespective of the concentration of magnetic atoms.

It is clear that the macroscopic magnetic properties of these alloys depends intimately upon the spatial distribution of atoms in the crystals; that is, on the short range atomic order. This fact is particularly apparent from the drastic changes in the magnetic susceptibility upon heat treatment. For example, in $\text{Cu}_{75}\text{Mn}_{25}$ the height of the cusp in the low field susceptibility is a factor of 4 larger in an annealed sample than in a quenched sample (9). It is known from neutron scattering experiments that the short range atomic order increases with annealing (10). This suggests that short range atomic order, or inhomogeneity, may be a prerequisite to spin glass behavior.

EXPERIMENTAL

The microscopic connection between the short range atomic order and the Mn spin-spin correlations can be observed using elastic neutron scattering in which the nuclear and magnetic scattering cross sections are separated by polarization analysis. Experiments of this type have previously been carried out on quenched, polycrystalline Cu-Mn samples, (11,7) yielding conflicting results. Ahmed and Hicks (11) concluded that the nearest neighbor Mn-Mn interaction is ferromagnetic, while the more recent results of Davis, et al (7), have been analyzed to yield antiferromagnetic coupling at nearest neighbor distances.

In order to clarify this situation we have carried out polarization analysis experiments at the HFIR reactor at Oak Ridge on a series of annealed Cu-Mn single crystal samples grown by the Bridgeman technique. The incident beam of wavelength 1.067 Å and polarization 99% was produced by an Fe (Si) monochromating crystal. The Cu-Mn crystals were placed at the sample position of the triple-axis spectrometer in a horizontal field of about 200 gauss directed along the scattering vector. The scattered neutrons were then analyzed by a Co(Fe) crystal. The polarization state of the incident beam was controlled by an rf flipper coil, thus permitting a measurement of the spin-flip $(\frac{d\sigma}{d\Omega})_{\uparrow\downarrow}$ and the non-flip $(\frac{d\sigma}{d\Omega})_{\uparrow\uparrow}$ cross sections separately (12). In this geometry, all of the magnetic scattering is spin flip scattering,

while the coherent nuclear scattering is non-spin flip scattering.

The Cu-Mn crystals were oriented with the [001] fcc axis normal to the scattering plane. Peaks in the diffuse nuclear scattering $(\frac{d\sigma}{d\Omega})_{\uparrow\uparrow}$ were observed at the $[\frac{1}{2}, 1, 0]$ -type positions in reciprocal space in all of the samples (open circles in Fig. 1). This scattering has the symmetry of the $M = 1$ (Cu_3Au) long period superlattice as has been discussed previously (5). The magnetic diffuse scattering $(\frac{d\sigma}{d\Omega})_{\uparrow\uparrow}$ peaks at the $[\frac{1}{2} \pm \delta, 1, 0]$ -type positions (x's in fig. 1). The intensity of both the nuclear and magnetic diffuse scattering decreases rapidly with decreasing Mn concentration as shown by the results of scans along the $[1, 1, 0]$ line displayed in Fig. 2. As a function of increasing temperature, the peaks in the magnetic diffuse scattering decrease in intensity and broaden continuously. This behavior is shown for the 25% Mn and 15% Mn samples in figs. 3 and 4. The nuclear diffuse scattering changes very little over this temperature range.

In order to determine, more precisely, the positions in reciprocal space where the magnetic diffuse scattering peaks, we have also carried out unpolarized neutron scattering experiments, to take advantage of the increased intensity available. Knowing, from the polarization analysis experiments, that the nuclear scattering is essentially constant over the temperature range 8K-295K, and that the magnetic scattering decreases to near zero at a temperature just above room temperature, a subtraction of the unpolarized scattering cross section at 295K from the cross section at 8K yields the magnetic scattering cross section at low temperatures. The results of this procedure are shown in Fig. 5. The splitting of the magnetic diffuse peaks is observed to increase as the Mn concentration decreases. It is seen that the absolute magnitudes of the magnetic differential scattering cross sections (as obtained with a calibration to the incoherent scattering cross section of vanadium) for the polarized and unpolarized data are in reasonable agreement.

DISCUSSION

It is clear that the magnetic diffuse scattering is symmetry-related to the short range order nuclear scattering. We interpret the scattering along the line $[1, 1, 0]$ to be due to one of three possible orientations of short range ordered clusters. That is, the magnetic diffuse peaks at $[\frac{1}{2} \pm \delta, 1, 0]$ along this line arises from scattering from the same regions of the macroscopic crystal as those that give rise to the diffuse nuclear

scattering at $[\frac{1}{2}, 1, 0]$. Similar symmetry-related magnetic and nuclear scattering is observed along the two other equivalent directions, namely $[1, k, 0]$ and $[1, 1, h]$.

In view of the fact that the small angle neutron scattering from Cu-Mn alloys clearly shows the freezing of spin fluctuations, (8) it may appear surprising that this magnetic diffuse scattering displays no hint of a freezing transition at low temperatures. However, we believe that this data is precisely the information we have needed to justify the qualitative model which has emerged over many years and has guided our thinking about magnetism in these spin glass alloys (13). This model involves the formation of magnetic clusters at some temperature well above T_f , and an increase in their concentration and average moment as the temperature is lowered toward T_f . The fluctuations within and among these clusters slows down as T approaches T_f . Whether or not critical slowing down should be expected is not clear. We would like to suggest that the model magnetic clusters are, in fact, the short range order domains, and that the data of figs. 2, 3, 4 and 5 represent the Fourier decomposition of the magnetism within these regions. With this data as the basis, we are currently working on various models to obtain a physical picture of the magnetic structure of these clusters in real space. We are also pursuing both inelastic and small angle neutron scattering experiments on these single crystal samples.

REFERENCES

- (a) Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract W-4705-eng-26 with the Union Carbide Corporation.
- 1) S. Valentiner and G. Becker, Z. Physik 80, 735 (1933).
- 2) E. Scheil and E. Wachtel, Z. Metallkde. 48, 571 (1957).
- 3) J. S. Kouvel, J. Phys. Chem. Solids 21, 57 (1961).
- 4) D. Meneghetti and S. S. Sidhu, Phys. Rev. 105, 130 (1957).
- 5) H. Sato, S. A. Werner, and M. Yessik, AIP Conference Proceedings, No. 5, (17th Annual Magnetism and Magnetic Materials Conference 1971)(1972).
- 6) P. Wells and J. H. Smith, J. Phys. F: Metal Physics 1, 763 (1971).
- 7) J. R. Davis, S. K. Burke and B. D. Rainford, J. Mag. and Mag. Mut. 15-18, 151 (1980).
- 8) A. P. Murani, Phys. Rev. Lett. 41, 1406 (1978); J. Mag. and Mag. Mat. (to be published) (1981).
- 9) See for example R. W. Tustian, Sol. State Comm. 19, 1075 (1976), and the reference given there.

- 10) S. A. Werner, H. Sato and M. Yessik, AIP Conference Proceedings, No. 10 (18th Annual Magnetism and Magnetic Materials Conference 1972) (1973).
- 11) N. Ahmed and T. J. Hicks, J. Phys. F5, 2168 (1975).
- 12) For a description of this spectrometer see R. M. Moon, T. Riste, and W. C. Koehler, Phys. Rev. 181, 920 (1969).
- 13) See ref. 3 and P. A. Beck, J. Less-Common Metals 28, 193 (1972).

Fig. 1. The (001) reciprocal lattice plane. The solid circles are the fcc Bragg points. The open circles are the positions of the peaks in the diffuse nuclear scattering and the x's are the locations of the peaks in the diffuse magnetic scattering.

Fig. 2. Lower half: The non-spin flip scattering cross section (nuclear diffuse scattering) measured along the line $[\lambda, \lambda, 0]$ of fig. 1. Upper half: The spin flip scattering cross section (magnetic diffuse scattering) measured along the line $[\lambda, \lambda, 0]$ of fig. 1. These data were taken at 8 K.

Fig. 3. Temperature dependence of the diffuse magnetic scattering for $\text{Cu}_{75}\text{Mn}_{25}$ along the $[2,1,0]$ line in reciprocal space.

$\text{Cu}_{75}\text{Mn}_{25}$

Fig. 4. Temperature dependence of the diffuse magnetic scattering for $\text{Cu}_{85}\text{Mn}_{15}$ along the $[2,1,0]$ line in reciprocal space.

Fig. 5. Magnetic diffuse scattering along the $[l, 1, 0]$ line in reciprocal space as measured with unpolarized neutrons at 8 K. (See text for a description of the procedure used to subtract the nuclear diffuse scattering.)