Moment clouds in *Cu*Mn

T. J. Hicks
Department of Physics, Monash University, Clayton, Victoria 3168, Australia

J. W. Cable
Solid State Division, ORNL, Oak Ridge, Tennessee

(Received 3 February 1997; revised manuscript received 18 May 1998)

Advantage was taken of the high magnetic susceptibility of Cu-Mn alloys in the range 12 to 22 at. % Mn to observe field-dependent magnetic neutron scattering from a single crystal containing 21.3 at. % Mn. The scattering was measured with unpolarized neutrons at low angles and between the (1,0,0) and $(1,\frac{1}{2},0)$ positions in reciprocal space using a neutron energy of 14.8 MeV. The magnetic scattering was approximately uniformly reduced by about 10% at 4.2 K after warming above the glass temperature and cooling again to 4.2 K in a field of 4.25 T. Reduction of the intensity of all magnetic features by the application of the field indicates that all the magnetic periodicities are intimately connected to the uniform magnetic response. A model which assumes that the alloy contains entities with both a ferromagnetic moment associated with the (0,0,0) scattering and an antiferromagnetic periodicity associated with the $(1,\frac{1}{2}\text{-}\delta,0)$ scattering yields a ferromagnetic moment for the entity of $36\mu_B$ estimated from the reduction in scattering. This compares with $42\mu_B$ estimated from the intensity and width of the low angle scattering. [S0163-1829(98)03934-4]

INTRODUCTION

Dilute solutions of manganese in copper have been regarded as the archetypal spin glass. Magnetic susceptibility studies[1] identified the hallmarks of spin glass behavior in the observation of a cusp in the susceptibility at a temperature associated with the spin freezing below which the susceptibility is a strong function of the thermal and magnetic history of the sample. A neutron diffraction experiment in which the magnetic cross section was definitely isolated using polarization analysis was that of Ahmed and Hicks.[2] The results of this experiment showed that the magnetic scattering below the glass temperature is overwhelmingly diffuse in nature with a tendency towards ferromagnetic spin correlations. Comparison with susceptibility measurements clearly indicates that at low temperatures the spatial fluctuation of the spins is almost entirely static with only a small proportion contributing to the susceptibility measured on a time scale of less than a minute. Later Gray, Hicks, and Smith[3] demonstrated that in more concentrated alloys the ferromagnetic correlation is strong and long ranged with ferromagnetic clusters with a radius of gyration of 26 Å.

In a series of papers[4,5] Werner, Cable, and others have identified broad satellite peaks in the magnetic cross section, again separated out using polarization analysis, which cluster around the $(1,\frac{1}{2},0)$ and equivalent reciprocal lattice positions and indicate an incommensurate modulation of the moment. These observations have lead Werner[6] to claim that Cu-Mn alloys are not spin glasses, but simply spin density waves (SDW's) as originally proposed by Overhauser in which the small domain size determines the breadth of the antiferromagnetic Bragg peaks near the $(1,\frac{1}{2},0)$ and similar positions, and the Fermi surface of the alloy determines the periodicity.

In the approach of Werner[6] the observed ferromagnetic correlations are regarded as separate from the SDW's and arising ''from the local ferromagnetic alignment of Mn spins within the ASRO (atomic short range order) clusters.'' Others[7,5] have attempted to explain the increase in magnetic scattering at small angles variously as due to genuine ferromagnetic regions within which there is an orthogonal antiferromagnetic periodicity giving rise to the magnetic satellite scattering in the $(1,\frac{1}{2},0)$ vicinity and also as an artifact of the scattering when there is an interference between the spin-spin and atom-atom correlations.[5]

The purpose of this experiment was to use the high susceptibility observed for alloys in the 12 to 22 at. % Mn range[8] with the application of a high field to modify the scattering. If the antiferromagnetic features in the scattering are unconnected with the high susceptibility, and therefore the ferromagnetic correlations, then those features should not be affected by the application of the field.

THE EXPERIMENT

The sample used in this experiment was a single crystal containing 21.3 at. % Mn which had been grown by the Bridgman technique and then quenched. The same crystal was used for a study of inelastic magnetic scattering by Tsunoda *et al.*[9]

The crystal was mounted with its [001] axis vertical in a cryostat with a split vertically aligned superconducting coil on the HB1 spectrometer at the HFIR reactor. The object was to measure the scattering near the forward direction and along the line in reciprocal space between (100) and $(1\frac{1}{2}0)$ at 4.2 K after cooling from above the glass temperature in both zero field and 42.5 kOe. All experiments used an incident neutron energy of 14.8 MeV and the spectrometer was set for elastic scattering.

The low angle experiments were performed with an incident collimation of 30 min and exit collimation of 20 min with the detector aperture height 5 cm at a distance of 150 cm from the specimen. The data covered the scattering

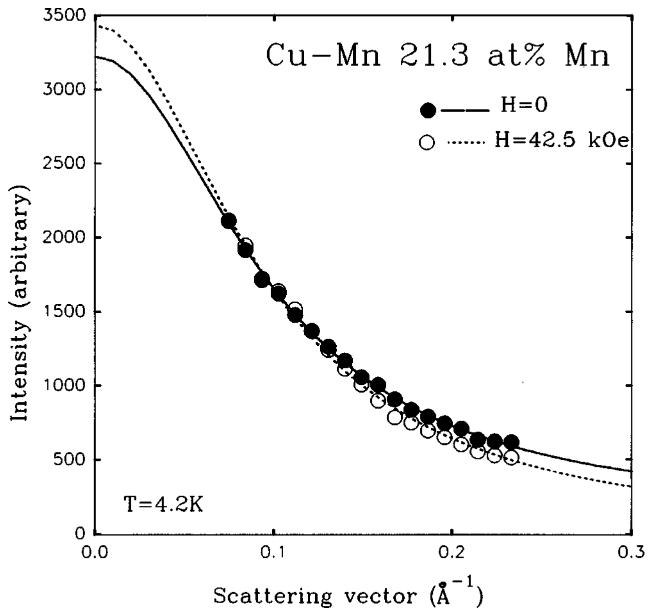

FIG. 1. The low angle scattering showing the increase towards small angles of the magnetic scattering and the effect of cooling in a field of 42.5 kOe.

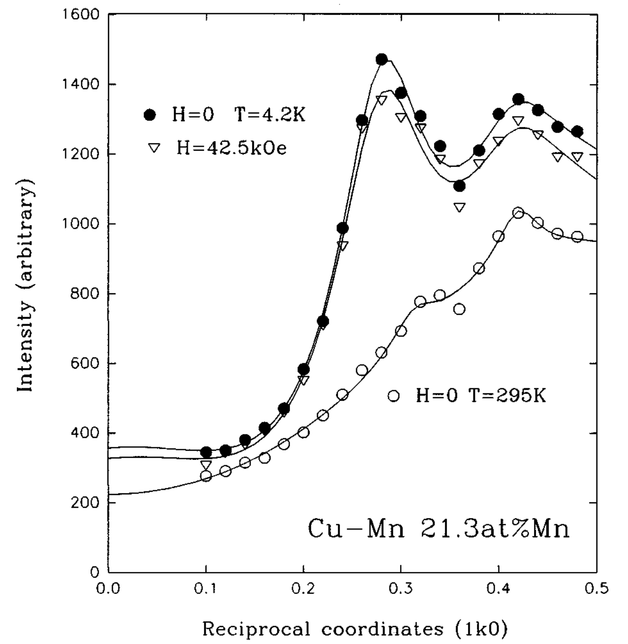

FIG. 2. The scattering between the (1,0,0) and $(1,\frac{1}{2},0)$ positions in reciprocal space for the zero field cooled and field cooled experiments. Also shown is the scattering in zero field at room temperature.

angles 1.6° to 5°. Figure 1 shows the scattering at low angles. Open points are taken in zero field and the closed points after cooling in 42.5 kOe. The lines are the fits of Lorentzians plus a background to the points. The backgrounds were within error of zero and the amplitude and half width of the fits are shown in Table I. It can be seen that at the larger scattering vectors the field cooled results are lower and, from the Lorentzian fits, that the half width of the field cooled scattering is less. The data themselves do not clearly show the reduction in half width which relies on the Lorentzian fitting. Attempts to use a Gaussian and a squared Lorentzian resulted in less good fits.

Further data was taken between the (1,0,0) and $(1,\frac{1}{2},0)$ reciprocal lattice positions, without altering the collimations, to encompass the nuclear and magnetic peaks identified by Cable *et al.*[5] Figure 2 shows the scan taken at room temperature and at 4.2 K after cooling from 130 K without a field and in a field of 42.5 kOe. The room temperature data shows a rise towards the $(1,\frac{1}{2},0)$ position which is predominantly due to nuclear defect scattering. Two small peaks are seen. The first is what remains at high temperature of the low-temperature magnetic satellite peaks. The second is clearly not affected by temperature and was also seen by Tsunoda, Kunitomi, and Cable[9] is possibly due to double Bragg scattering or a change in crystal attenuation.

At low temperature the magnetic satellites are seen along with a further increase in scattering towards the $(1,\frac{1}{2},0)$ position. Figure 2 shows the difference between the low-temperature results and the room-temperature run. The magnetic satellite peak is clearly visible, however, the other peak does not appear. Application of a field before cooling again reduces the magnetic scattering.

TABLE I. Fitted parameters for the low angle scattering.

Parameter	ZF cooled	F cooled in 42.5 kOe
$\kappa=0$ intensity	3106	3433
Half width	0.099 Å^{-1}	0.096 Å^{-1}

This data was fitted to a function describing the diffuse nuclear and magnetic scattering plus two Lorentzians, constrained to have the same width, for each of the two peaks superimposed. Gaussians for the peaks fitted less well. The function can be written

$$I=a_1+a_2\cos(2\pi ky)+a_3\cos(4\pi ky)$$

$$+\frac{a_4}{[a_5+(a_6-k)^2]}+\frac{a_7}{[a_5+(a_8-k)^2]}.$$

The fitted constants a_2 and a_3 are related to the Cowley short-range order parameters. These are the lowest two modulations which can be obtained from data along the $(1,k,0)$ direction because the (1,0,0) and (1,1,0) are equivalent points and the function must be even around each. The last two terms are the Lorentzians describing the two sharper peaks.

The parameters a_1, a_2, and a_3 largely describe the nuclear diffuse scattering although Cable *et al.*[5] show that part of the broad scattering around the $(1,\frac{1}{2},0)$ is magnetic. Nevertheless the fitted parameters (Table II) for the three sets of data at different temperatures and fields are broadly simi-

TABLE II. Raw fitted parameters relating to background (a_1) and short-range order parameters.

	295 K	4.2 K ($H=0$)	4.2 K ($H=4.25$ T)
a_1	470±50	500±60	440±100
a_2	−280±50	−380±50	−340±70
a_3	70±40	190±50	180±75

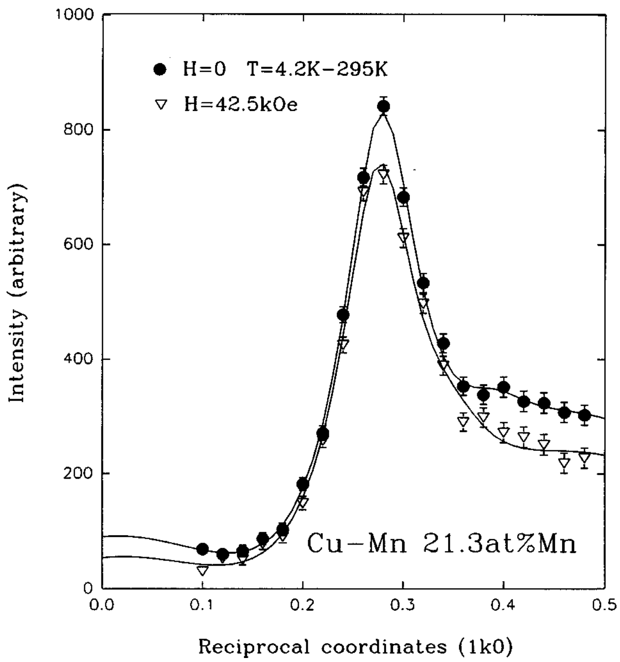

FIG. 3. The zero field cooled and field cooled scattering between (1,0,0) and $(1,\frac{1}{2},0)$ with the room-temperature scattering subtracted.

lar. This is in agreement with the Cable *et al.* results which show that the broad magnetic scattering around the $(1,\frac{1}{2},0)$ is virtually unchanged up to about room temperature for these Mn concentrations. By comparison of the intensity of our satellite peak, as shown in Fig. 3, with the Cable *et al.* Fig. 7, each count in a (1,0,0) to $(1,\frac{1}{2},0)$ scan is approximately 1 mb sr^{-1} at.$^{-1}$. The parameters are therefore roughly in units of mb sr^{-1} at.$^{-1}$. The average nuclear defect cross section for this composition, calculated from the scattering lengths is 210 mb sr^{-1} at.$^{-1}$ so that it can be seen that the average contributions to the cross section from magnetic and nuclear sources are comparable. The nuclear part of the fitted a_2 is related to a sum of all those Cowley short-range order parameters which contain a modulation with a wavelength equal to the lattice parameter along y^5. This includes the second neighbor short-range order parameter α_{020}, that for third neighbors α_{121}, as well as fourth α_{022} and α_{220}, and all other SRO parameters with $m=2$. $a_2=c(1-c)\Sigma_n A_{2n} e^{in}$ with $A_{2n}=\Sigma_l \alpha_{lnm}$ as defined by Cable *et al.* Using the few values for A_{2n} determined for a 25 at. % crystal by Cable *et al.* gives $a_2=-100$ for the nuclear part of the scattering. There is also considerable modulation of the magnetic part of the scattering which will contribute to a_2 and the set of A_{2n} used is not complete. On the other hand, we might expect smaller SRO parameters for a less concentrated sample. Still the parameters obtained are reasonable enough to have confidence in this part of the fitting function. The Lorentzian fit to the $(1,\frac{1}{2}\text{-}\delta,0)$ peak gave a width of 0.048 reciprocal lattice units or 0.082 Å^{-1} which is similar to the width of the low angle peak.

DISCUSSION

The experiment clearly shows that application of a magnetic field affects the scattering associated with the antiferromagnetic correlations. There are two possibilities. Either the field directly affects the antiferromagnetic correlations or they are rigidly connected to the ferromagnetic component which responds to the field. If the former is correct the most likely modification is that moments in small exchange fields near the nodes of the SDW's respond to the field and distort the simple form of the SDW's. To describe the resulting form of the SDW's requires that other Fourier components be added to the zero field single Fourier component. Thus the effect of this, the most likely modification of the antiferromagnetic correlations, is not a reduction of the antiferromagnetic features but an addition of scattering at other scattering vectors. Arrott[10] has also suggested that to maximize the number of moments which can respond to the field the phase of the SDW's may be shifted in an applied field. This does not change the amplitude of the SDW's and is probably of minimal importance as the Mn moments are randomly distributed along the SDW's.

The second possibility can only occur if the antiferromagnetic correlations are intimately connected to the ferromagnetic correlations which produce the high susceptibility above the freezing temperature. The simplest hypothesis to explain this result is that the two periodicities are rigidly connected. If this were the case entities incorporating both periodicities would respond to the field and in addition scatter independently as superparamagnetic particles. Each would have a spin distribution, incorporating the two periodicities, which could be described with an entity form factor. This model has the virtue that both features of the form factor, ferromagnetic and antiferromagnetic, would respond to the field in the same manner. That this is not exactly the case can be seen from the low angle results where the scattering is not reduced to the same extent by the application of the field. In fact there may be a crossover at the low limit of our results. In the context of scattering from individual entities this means that the form factor cannot be absolutely rigid. Nevertheless the assumption of a rigid entity form factor seems a good starting point to see whether a superparamagnetic entity based model can explain the results especially as the widths of the peaks from the ferromagnetic and antiferromagnetic correlations are so similar suggesting that both widths are limited by the entity size.

The scattering cross section from a paramagnetic system in the quasielastic approximation is

$$\frac{d\sigma}{d\Omega}=\left(\frac{e\gamma}{\hbar c}\right)^2[\chi_{xx}(\kappa)+\chi_{zz}(\kappa)]k_B T$$

in which $\chi(\kappa)$ is the wave-vector-dependent susceptibility along x and z with the scattering vector κ along the orthogonal Cartesian direction y. The other symbols have their usual meaning.

If we assume that the paramagnetic susceptibility is due to superparamagnetic Langevin entities we can write

$$\chi(\kappa)=\frac{N\mu^2 f^2(\kappa)}{3k_B T}$$

at zero field, where μ is the entity moment and $f(\kappa)$ is its form factor assumed to contain both the ferromagnetic and antiferromagnetic correlations between individual manganese moments. When a field is applied along the z direction the appropriate z component of the susceptibility is the *differential* susceptibility at that field. So

$$\chi_{zz}(\kappa)=\left(\frac{dM_z(\kappa)}{dH}\right)_H$$

with the Langevin wave-vector-dependent magnetization

$$M_z(\kappa)=N\mu f(\kappa)\left[\coth\left(\frac{uH}{k_BT}\right)-\frac{k_BT}{\mu H}\right].$$

This gives

$$\chi_{zz}(\kappa)=\frac{N\mu^2 f^2(\kappa)}{k_BT}\left[\frac{1}{\xi^2}-\operatorname{cosech}^2\xi\right]$$

with $\xi=\mu H/k_BT$. For large fields and moments this will be considerably less than the zero field susceptibility leading to the reduction in cross section observed.

The observed reduction in cross section is about 10%. However, this must be due entirely to $\chi_{zz}(\kappa)$ as the contribution to the cross section from $\chi_{xx}(\kappa)$ is unaltered. A 20% reduction in $\chi_{zz}(\kappa)$ corresponds to about $\xi=1$. If we then assume that the characteristic temperature is approximately the glass temperature $T_g=100$ K, we can get an estimate for the size of the moment from $\mu\approx k_BT/H$. This gives $36\mu_B$ per entity.

From the half width of the forward peak Γ, the $1/e$ radius of the ferromagnetic correlation is about 10 Å or a volume containing about 90 cubic unit cells. The forward scattering cross section can be roughly estimated from previous experiments such as that by Cable *et al.*[5] By comparison we find that one count in this experiment is roughly equal to 1 mb sr^{-1} at.$^{-1}$ in the (1,0,0) to $(1,\frac{1}{2},0)$ scans. Transferring this to the low angle scans gives a forward cross section of about 3.3 b sr^{-1} at.$^{-1}$. The forward cross section is given by

$$\frac{d\sigma}{d\Omega}=\frac{2}{3}x\mu^2,$$

where there are x entities with total moment μ. Using $x=0.0028$ from the approximately 360 atoms associated with each entity we calculate $\mu=42\mu_B$ or about $0.6\mu_B$ per Mn atom. This is satisfying as the two estimates of entity moment, one based entirely on the reduction of cross section due to field cooling and the other based entirely on the magnitude and width of the forward scattering, give similar results.

CONCLUSION

This experiment clearly shows that the ferromagnetic and the antiferromagnetic correlations are intimately connected to the uniform magnetic response. In other words, the ferromagnetic and antiferromagnetic regions are not spatially separate as in some previous models.

The superparamagnetic entity model introduced here is a first attempt to extract more than a qualitative understanding of the field and temperature behavior of the magnetic scattering. It is based on the observation that to a first approximation the magnetic intensity reduction observed is independent of scattering vector and yields reasonable results for superparamagnetic entity size and moment. It is not the last word particularly because extrapolation of the results towards small scattering vectors would suggest that the magnetic scattering there is enhanced by the application of the field.

The results do not define the actual spin configuration of this superparamagnetic entity but a ferromagnetically distorted spin density wave (SDW) is certainly consistent. Such ferromagnetically distorted SDW's occur, for example, in the rare-earth metals in applied fields. These are the well known fan and helifan structures that are static and long ranged as opposed to the dynamic and short-ranged CuMn order but similar local spin ordering may occur in both systems.

ACKNOWLEDGMENTS

ORNL is managed by Lockheed Martin Energy Research under Contract No. DE-AC05-96OR22464 for the U.S. Department of Energy. T.J.H. is grateful for the hospitality of the Solid State Division of ORNL for the duration of this experiment.

[1] S. Nagata, P. H. Keesom, and H. R. Harrison, Phys. Rev. B **19**, 1633 (1979).

[2] N. Ahmed and T. J. Hicks, Solid State Commun. **15**, 415 (1974).

[3] E. M. Gray, T. J. Hicks, and J. H. Smith, J. Phys. F **12**, L189 (1982).

[4] S. A. Werner and J. W. Cable, J. Appl. Phys. **52**, 1757 (1981); J. W. Cable, S. A. Werner, G. Felcher, and N. Wakabyashi, Phys. Rev. Lett. **49**, 829 (1982).

[5] J. W. Cable, S. A. Werner, G. Felcher, and N. Wakabyashi, Phys. Rev. B **29**, 1268 (1984).

[6] S. A. Werner, Comments Condens. Matter Phys. **15**, 55 (1990).

[7] T. M. Harders, T. J. Hicks, and J. H. Smith, J. Phys. F **13**, 1263 (1983).

[8] P. A. Beck, J. Less-Common Met. **28**, 193 (1972).

[9] Y. Tsunoda, N. Kunitomi, and J. W. Cable, J. Appl. Phys. **57**, 3753 (1985).

[10] A. S. Arrott, in *Magnetism IIB*, edited by G. T. Rado and H. Suhl (Academic, New York, 1966), pp. 384–396.