

lower magnetic fields, that is, at fields below the onset of the dHS oscillations. We do not observe any evidence for 1D spin dynamics in the low-field regime.

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Neutron-Polarization-Analysis Study of the Spin Structure of Cu-Mn Spin-Glasses

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Neutron-polarization-analysis measurements on single-crystal Cu-Mn alloys show the presence of a long-period spin modulation with a concentration-dependent period. Coexistence with these modulated regions are smaller regions in which the spin correlations are determined by the atomic short-range order and which have ferromagnetic moments. The interactions between these ferromagnetic and modulated regions are an essential element in understanding the complicated magnetic behavior of this spin-glass system.

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The peculiar magnetic behavior¹ of Cu-Mn alloys, and that of other spin-glass systems, is generally attributed to the interactions between spatially inhomogeneous ferromagnetic-antiferromagnetic regions. Despite intense experimental and theoretical effort over the past few years, fundamental questions regarding the spin-glass state remain unresolved. Part of the difficulty in developing an adequate theory is a lack of information on the microscopic spin structure of a spin-glass. Inelastic neutron data² on Cu-Mn alloys indicate ferromagnetic clusters that fluctu-

ate in time while quasielastic neutron diffuse scattering data^{3,4} yield short-range order (SRO) parameters that are antiferromagnetic for first neighbors and ferromagnetic for second neighbors. In this paper, we describe recent neutron-polarization-analysis data which show that the spin correlations in Cu-Mn are more complicated than was previously observed.^{3,4} There are long-wavelength modulations that persist over large distances coexisting with ferromagnetic correlations over shorter distances which are directly associated with the atomic SRO. The interactions be-

tween these small ferromagnetic regions and the larger modulated regions are undoubtedly an essential ingredient in understanding the unusual magnetic behavior of this spin-glass system.

The nuclear and magnetic cross sections were separated by the polarization analysis method⁵; these are illustrated in Fig. 1 where the data for the 25% alloy at 10 K are presented as intensity contours in an (010) reciprocal-lattice plane.

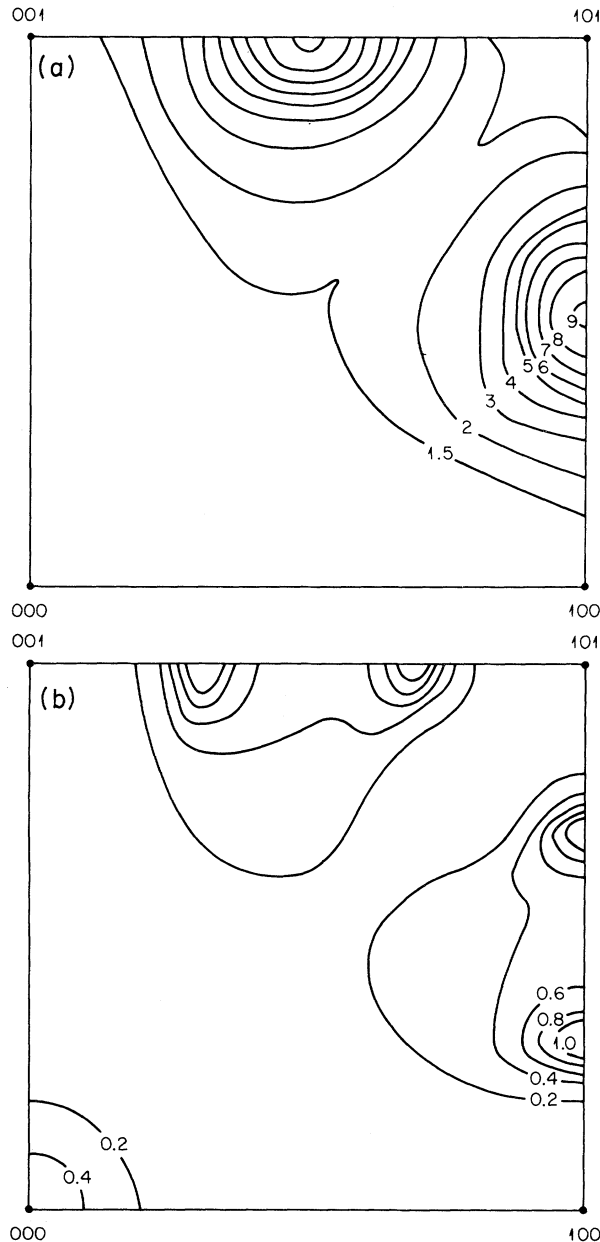


FIG. 1. The (a) nuclear and (b) magnetic scattering from Cu-(25 at.% Mn) at 10 K. Intensity contours are in $S(\vec{k})$ units in (a) and in barns steradian⁻¹ atom⁻¹ in (b).

The nuclear scattering shown in Fig. 1(a) exhibits broad diffuse peaks at $(1, 0, \frac{1}{2})$ and at other symmetry-related positions. The magnetic scattering shown in Fig. 1(b) has intensity distributed around $(0, 0, 0)$ and $(1, 0, \frac{1}{2})$ but the most prominent features are the relatively sharp peaks at $(1, 0, \frac{1}{2} \pm \delta)$ and other symmetry-related positions. The positions of these peaks indicate a long-period modulation of the spin correlations and their widths show that this modulation persists over appreciable distances of about $10a_0$. With increasing temperature these peaks become broader and less intense with no apparent change in peak positions. The spin correlations vanish at a concentration-dependent temperature much higher than T_f with no apparent discontinuity at T_f . Similar intensity distributions are observed for the less-concentrated alloys. At all concentrations, the distribution along $[10l]$ is resolvable into two relatively sharp peaks at $(1, 0, \frac{1}{2} \pm \delta)$ and a Gaussian peak centered at $(1, 0, \frac{1}{2})$ with the same shape as the nuclear peak. This shows that short-range spin correlations coexist with the long-period modulations. A homogeneous coexistence is suggested by the concentration dependence of the magnetic intensity at $(1, 0, \frac{1}{2} \pm \delta)$ which is proportional to c^2 .

The spin-nonflip or nuclear cross section is given by

$$\frac{d\sigma^{++}}{d\Omega}(\vec{k}) = c(1-c)(b_{\text{Mn}} - b_{\text{Cu}})^2 S(\vec{k}), \quad (1)$$

where $b_{\text{Mn}} = -0.373$, $b_{\text{Cu}} = 0.772 \times 10^{-12}$ cm, and

$$S(\vec{k}) = \sum_{\vec{r}} \alpha(\vec{r}) \exp(i\vec{k} \cdot \vec{r}), \quad (2)$$

where the $\alpha(\vec{r})$ are Warren-Cowley SRO parameters. The atomic SRO in this system can be regarded as a damped composition-modulation wave with wave-vector $\vec{Q}_N = 2\pi(\vec{b}_1 + \frac{1}{2}\vec{b}_3)$. This describes a z -axis modulation with a period of $2a_0$ and with unlike nearest neighbors in the x - y plane. Equivalent tetragonal domains develop along the x and y axes producing an intensity distribution with cubic symmetry. The data of Fig. 1(a) were Fourier transformed to obtain the $\alpha(\vec{r})$ averaged over domains. These indicate that Mn atoms prefer Cu first neighbors and Mn second neighbors. Since first neighbors presumably interact antiferromagnetically and second neighbors interact ferromagnetically, SRO should enhance the ferromagnetic properties of the system. There is abundant experimental evidence^{6,7} that this is indeed the case.

The paramagnetic cross section is inelastic but

the observed² energy transfers are only about 1 meV. In this experiment, the incident energy was 72 meV and the energy resolution was 2.5 meV half width at half maximum so that the quasielastic approximation should be valid. In this approximation, the paramagnetic cross section is

$$\frac{d\sigma^{-+}}{d\Omega}(\vec{K}) = \left(\frac{e^2\gamma}{mc^2}\right)^2 f(\vec{K})^2 \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \hat{K}_\alpha \hat{K}_\beta) \sum_{\vec{R}} \langle S_{\vec{R}}^\alpha S_0^\beta \rangle \exp(i\vec{K} \cdot \vec{R}), \quad (3)$$

where $f(\vec{K})$ is the magnetic form factor, α and β denote Cartesian components of the spin, and $\langle S_{\vec{R}}^\alpha S_0^\beta \rangle$ is the quasistatic spin-spin correlation.

The magnetic cross sections cannot be simply Fourier transformed to obtain magnetic SRO parameters because of the long correlation lengths associated with the $(1, 0, \frac{1}{2} \pm \delta)$ peaks. Instead, we calculate the cross sections for assumed spin-correlation models and compare directly with the observed cross sections. The data show that the magnetic and nuclear intensities at $(1, 0, \frac{1}{2})$ are proportional to each other. This shows that the spin correlations have the same signs, range, and relative magnitudes as the atom-pair correlations. If $p_{\vec{R}}$ is the number of Mn atom at \vec{R} , then the Mn-Mn pair correlation is $\langle p_{\vec{R}} p_0 \rangle = c^2 + c(1-c)\alpha(\vec{R})$. For a composition modulation, this fluctuates about c^2 between the limits of $2c^2$ and zero. Since the collinear Mn-Mn spin correlation must fluctuate about zero between the limits of $+S_{Mn}^2$ and $-S_{Mn}^2$, the Mn-Mn spin correlation for $\vec{R} \neq 0$ can be written as

$$\langle S_{Mn}(\vec{R}) S_{Mn}(0) \rangle = S_{Mn}^2 (\langle p_{\vec{R}} p_0 \rangle / c^2 - 1). \quad (4)$$

Since the Cu atoms have no magnetic moments, this must be multiplied by $\langle p_{\vec{R}} p_0 \rangle$ to obtain the total spin correlation in Eq. (3). The cross section for z -axis domains with the spins in the x - y plane is then

$$\frac{d\sigma^{-+}}{d\Omega}(\vec{K}) = \left(\frac{e^2\gamma}{mc^2}\right)^2 f(\vec{K})^2 \frac{1+R_z^2}{2} S_{Mn}^2 [c(1-c)S(\vec{K}) + (1-c)^2 T(\vec{K}) - (1-2c)], \quad (5)$$

where $S(\vec{K})$ is defined by Eq. (2) and

$$T(\vec{K}) = \sum_{\vec{R}} \alpha^2(\vec{R}) \exp(i\vec{K} \cdot \vec{R}). \quad (6)$$

The $S(\vec{K})$ term is the same as in the nuclear cross section and peaks at $\vec{K} = \vec{G} \pm \vec{Q}_M$, where \vec{G} is a fundamental reciprocal-lattice vector, whereas the $T(\vec{K})$ term peaks at $\vec{K} = \vec{G}$ and is broader and less intense than $S(\vec{K})$. This peaking at the fundamental positions reflects the ferromagnetic character of these SRO regions, which results from the spin-pair and atom-pair correlations being in phase. S_{Mn} values extracted from the observed magnetic and nuclear cross sections at $(1, 0, \frac{1}{2})$ by use of Eq. (5) are given in Table I. These are near the expected value of $S = 2$ at all concentrations.

In considering the peaks that arise from the long-period modulations, we note that the data do not define the type of modulation. This could be sinusoidal, as in a spiral configuration, or square wave, as in an antiphase domain. The scattering from these and from other intermediate type modulations is quite similar. In the following calculation, we use a spiral model for simplicity. The data show that there is a long-period modulation with a wave vector of $\vec{Q}_M = 2\pi [\tilde{b}_1 + (\frac{1}{2} - \delta)\tilde{b}_3]$. This corresponds to a modulation along the z axis with a period of $a_0/(\frac{1}{2} - \delta)$ and with the condition that first-neighbor spins in the x - y plane are oppositely aligned. Equivalent tetragonal domains develop along the x and y axes.

The cross section for a spiral configuration with the spins in the x - y plane normal to the tetragonal z axis is

$$\frac{d\sigma^{-+}}{d\Omega}(\vec{K}) = \left(\frac{e^2\gamma}{mc^2}\right)^2 f(\vec{K})^2 \frac{1+\hat{K}_z^2}{4} S_{Mn}^2 \{c^2 \exp[i(\vec{K} \pm \vec{Q}_M) \cdot \vec{R}] + c(1-c)S(\vec{K} \pm \vec{Q}_M)\}, \quad (7)$$

where the first term gives Bragg peaks at $\vec{K} = \vec{G} \pm \vec{Q}_M$ and the second term gives broad peaks at $\vec{K} = \vec{G} \pm \vec{Q}_M \mp \vec{Q}_M$. Of course, Bragg peaks are not observed because the spiral order persists only over distances of about $10a_0$. Nevertheless, the volume integrals of the calculated and observed peaks can be compared to obtain the S_{Mn} values listed in Table I. These are approximately the same at each concentration but are smaller than those derived from the ferromagnetic SRO. This is not surprising because the spiral order is locally perturbed by the ferromagnetic regions. This perturbation is not enough to destroy the coherence of the spiral but would decrease the peak intensities of the spiral.

TABLE I. Magnetic parameters for Cu-Mn alloys.

c	$\frac{1}{2} - \delta$	S_{Mn}^a	S_{Mn}^b
0.05	0.16	1.94	1.03
0.10	0.21	1.82	1.07
0.15	0.25	1.96	1.04
0.25	0.31	1.67	1.19

^aFrom $(1, 0, \frac{1}{2})$ intensity and Eq. (5).

^bFrom $(1, 0, \frac{1}{2} \pm \delta)$ intensity and Eq. (7).

The second term in Eq. (7) distributes intensity into diffuse peaks displaced by $0, 0, \pm\delta$ from each \vec{G} . These peaks have the same half-width as $S(\vec{K})$ which is approximately the same as their displacement from \vec{G} . The x and y domains also produce such peaks displaced by $\pm\delta, 0, 0$ and $0, \pm\delta, 0$ and the net result is a broad peak centered at each \vec{G} . At (000) for the 25%-Mn alloy, we calculate 0.47 b arising from this $S(\vec{K} \pm \vec{Q}_M)$ term. An additional 0.14 b is contributed by the terms in Eq. (5). The total of 0.61 b compares favorably with the "observed" value of 0.5 b obtained by extrapolation of an $[h00]$ scan.

Thus, the assumed spin correlations account for all of the observed magnetic scattering. The $(1, 0, \frac{1}{2} \pm \delta)$ peaks are from a long-period modulation and the broad peaks at $(1, 0, \frac{1}{2})$ arise from short-range spin correlations that are directly associated with the atom-pair correlations. It is only the convolution terms between the spin and atomic correlations which give rise to the diffuse peaks at \vec{G} .

The microscopic spin structure for Cu-Mn alloys that emerges from these considerations is basically a long-period modulation. This is not a simple modulation in the sense that antiparallel alignment is required for nearest-neighbor spins in the plane normal to the modulation direction. The period of the modulation is concentration dependent and varies from approximately $6a_0$ at 5% Mn down to about $3a_0$ at 25% Mn. Although long-range order does not develop, this modulation occurs over large volumes in real space with dimen-

sions of about $10a_0$. According to the model calculations, this is the only type of spin order that would occur if these were random alloys. However, additional spin correlations develop in the presence of atomic SRO. These short-range spin correlations have the same signs and range as the atomic correlations. For Mn-Mn pairs these produce fewer than average Mn first neighbors, which align antiparallel, and more than average Mn second neighbors, which align parallel. This results in ferromagnetic regions with dimensions defined by the range of the atomic SRO. These small ferromagnetic regions coexist homogeneously with the much larger modulated regions where they locally perturb the modulation without destroying its coherence. We suggest that the magnetic behavior of this system, and probably that of similar systems such as Au-Mn, Ag-Mn, Pd-Mn and Ni-Mn, is determined by the magnitude and temperature dependence of the interactions between these small ferromagnetic regions and the larger modulated regions.

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