conservative estimate of the error in these values of the transition temperature is $\pm 0.03J$. Alternatively, of course, if our values for the transition temperatures are correct, then the MC results confirm the KT prediction for $K_\infty(T_R)$ for these systems. Finally, we mention that we have also studied dynamic correlation functions and find results in very good agreement with the theory of Chui and Weeks.[9] Further discussion of these results will appear in a forthcoming publication. We are grateful to R. J. Myerson, P. C. Hohenberg, and S. T. Chui for helpful discussions.

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Neutron-Scattering Measurement of the Edwards-Anderson Order Parameter for a *Cu*-Mn Spin-Glass Alloy

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We have carried out neutron inelastic scattering measurements on a Cu–8-at.%-Mn alloy employing different energy resolutions varying over two orders of magnitude. The temperature dependence of the elastic magnetic scattering cross section which is related to the Edwards-Anderson order parameter is found to vary systematically with the energy resolution employed to separate the elastic scattering from the inelastic/quasielastic contributions and hence on the effective time constant of measurement.

Recent phase-transition theories of spin-glasses predict an "ordered" magnetic phase characterized by an order parameter $q = [\langle S_i \rangle^2]_{av}$ which vanishes at a well-defined transition temperature T_f.[1] The theoretical situation, however, is not entirely clear. In particular, using the high-temperature series-expansion techniques, Fisch and Harris[2] have shown that the susceptibility exponent associated with the Edwards-Anderson (EA) order parameter increases with decreasing dimensionality below $d = 6$ and diverges at $d = 4$, suggesting that if a phase transition in real three-dimensional spin-glasses occurs it may not be related to the critical behavior of the EA order parameter.

In the following we outline how the EA order parameter for an ideal spin-glass is formally related to the elastic-neutron-scattering cross section[3] and we report on our attempts to measure it for a Cu–8-at.%-Mn alloy using spectrometers with high elastic energy resolutions varying over two orders of magnitude. An important conclusion which may be drawn from the results is that the true EA order parameter is not accessible to any practical probes employing finite measurement-time constants because of the wide spectrum of relaxation times of the spin system extending to infinity at temperatures around and below that of the peak in the magnetic susceptibility.

The double-differential neutron-scattering

cross section for a spin system can be expressed as[4]

$$\frac{d^2\sigma}{d\Omega\, d\omega} = N\left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k_0} S(Q,\omega), \tag{1}$$

where

$$S(Q,\omega) = (N\pi)^{-1} F^2(Q) \sum_{ij} \int_{-\infty}^{\infty} \exp[i\vec{Q}\cdot(\vec{r}_i - \vec{r}_j) - i\omega t] \langle S_i(0) S_j(t)\rangle\, dt\,, \tag{2}$$

and the symbols have their usual meaning. The pair-correlation function can be separated into two parts:

$$\langle S_i(0) S_j(t)\rangle = \{\langle S_i(0) S_j(t)\rangle - \langle S_i\rangle\langle S_j\rangle\} + \langle S_i\rangle\langle S_j\rangle, \tag{3}$$

where the first term inside the curly brackets is purely inelastic/quasielastic and the second term is elastic. The latter gives

$$S_e(Q) = 2N^{-1} F^2(Q) \sum_{ij} \exp[i\vec{Q}\cdot(\vec{r}_i - \vec{r}_j)] \langle S_i\rangle\langle S_j\rangle = 2N^{-1} F^2(Q) \sum_{in} \exp[-i\vec{Q}\cdot\vec{r}_n] \langle S_i\rangle\langle S_{i+n}\rangle, \tag{4}$$

where $n = j - i$ and $\vec{r}_n = \vec{r}_j - \vec{r}_i$. The summation over i yields

$$S_e(Q)/2F^2(Q) = \sum_n \exp[-i\vec{Q}\cdot\vec{r}_n] [\langle S_i\rangle\langle S_{i+n}\rangle]_{\mathrm{av}},$$

where $[\,]_{\mathrm{av}}$ indicates an average over all spins. Taking the inverse Fourier transform of the above equation yields the following:

$$N^{-1} \sum_Q \exp[i\vec{Q}\cdot\vec{r}_m] S_e(Q)/2F^2(Q) = [\langle S_i\rangle\langle S_{i+m}\rangle]_{\mathrm{av}}. \tag{5a}$$

Hence for $m = 0$ we obtain the order parameter q defined by EA

$$q = [\langle S_i\rangle\langle S_i\rangle]_{\mathrm{av}} = N^{-1} \sum_Q S_e(Q)/2F^2(Q). \tag{5b}$$

For an ideal spin-glass the quantity $S_e(Q)/2F^2(Q)$ is independent of Q so that measurement of the elastic scattering at any Q value yields directly the EA order parameter. In real spin-glasses, containing a-few-percent solute magnetic atoms, short-range magnetic correlations are always present despite complete positional randomness of the two species of atoms. Hence, the measurement of the EA order parameter involves the determination of $S_e(Q)$ over the whole Q range which is, of course, not always possible to do. Measurement of the elastic scattering over a limited Q range can, nevertheless, yield information which reflects the real behavior of the order parameter.

The determination of the purely elastic scattering, however, is limited by the instrumental energy resolution. In conventional measurements even the coarse energy resolution of standard spectrometers is quite adequate to separate the elastic scattering from inelastic processes. For the spin-glass problem, however, the question of the energy resolution is of special importance because of the wide spectrum of relaxation times of the spin system, evident in many of its physical properties.[5] Computer simulation experiments[6] show how the self-correlation function for the spins evolves from one showing a rapid exponential decay at high temperatures to that with very slow, complex decay form at lower temperatures. We may therefore express the self-correlation function in the form valid at all temperatures:

$$\langle S_i(0) S_i(t)\rangle = \sum_{\tau_\alpha} P(T,\tau_\alpha) \exp(-t/\tau_\alpha), \tag{6}$$

where $P(T,\tau_\alpha)$ is the probability distribution of relaxation time τ_α at temperature T.

For a relaxation process with time constant τ_α to be resolved in the conventional neutron-scattering experiment, the energy resolution ΔE of the spectrometer should be at least comparable to $\sim\hbar/\tau_\alpha$; if not, the scattering would be indistinguishable from that due to truly elastic processes. A neutron spectrometer with energy resolution ΔE_0 can therefore resolve processes with the longest relaxation times τ_0 given by $\tau_0 \sim \hbar/\Delta E_0$, and the rest with times longer than this appear as elastic scattering. Hence the latter is proportional to the quantity q_{τ_0} given by

$$q_{\tau_0} = [\langle S_i\rangle_{\tau_0}{}^2]_{\mathrm{av}} = \sum_{\tau_\alpha > \tau_0}^{\infty} P(T,\tau_\alpha) \exp(-t/\tau_\alpha), \tag{7}$$

where the summation is over all relaxation times longer than τ_0, the limit set by the instrumental energy resolution ΔE_0.

Another probe of the local magnetization $[|\langle S_i\rangle_{\tau_0}|]_{\mathrm{av}}$ and hence the "order parameter" $q_{\tau_0} = [|\langle S_i\rangle_{\tau_0}|^2]_{\mathrm{av}}$ is the Mössbauer effect. In this case the local magnetization $\langle S_i\rangle$ produces a hyperfine field at the nucleus which leads to the splitting of the γ-ray spectrum of suitable radioactive nuclei. For the splitting to be observed, however, it must be comparable to or larger than the natural width of the Mössbauer line giv-

en by its lifetime τ_0. Also the local magnetization $\langle S_i \rangle$ need remain stable only over the corresponding period τ such that $\tau \gtrsim \tau_0$ which therefore determines the time constant of the measurement ($\sim 10^{-7}$ s for Fe^{57}). Hence, in a Mössbauer experiment one measures

$$q_{\tau_0}^{1/2} = [|\langle S_i \rangle_{\tau_0}|]_{av} = N^{-1} \sum_{\tau_\alpha > \tau_0} |\langle S_i \rangle_{\tau_\alpha}| ,$$

where τ_0 is the intrinsic lifetime of the Mössbauer transition. Thus, the determination of the order parameter by this technique is also limited by the finite time constant of measurement.

We may recall that in addition to the magnetic scattering discussed above there is also a nonmagnetic contribution to the elastic scattering. This arises from the nuclear incoherence due to the binary mixture as well as from the incoherence of each of the two species of atoms. For a random substitutional alloy this gives a Q-independent scattering cross section except for the Q dependence through the Debye-Waller factor. This nonmagnetic contribution to the elastic scattering can be distinguished from the magnetic scattering because the former is almost temperature independent (the temperature dependence due to the Debye-Waller factor being relatively small below room temperature), and the magnetic scattering at temperatures well above the freezing temperature is purely inelastic (quasielastic with a large width) and lies outside the elastic energy resolution of the instruments used. Hence, the temperature-dependent part of the elastic scattering represents purely the magnetic contribution.

The first set of measurements were made on the multichopper time-of-flight (TOF) spectrometer IN5 with neutrons of incident energy 3.0 meV and an elastic energy resolution of ~230 μeV.[7] We have also carried out measurements using the backscattering spectrometer IN10 over a small energy window of ± 12 μeV and an elastic resolution of ~1.5 μeV using neutrons of incident energy 2.1 meV. The observed spectra were corrected for background scattering self-shielding and absorption, and normalized with respect to scattering obtained from a vanadium sample. Multiple-scattering contributions which give a constant correction factor were estimated to be less than 5% and were not included. It should be noted that in the backscattering technique the "flat background" is a function of the total scattering cross section of the sample so that small systematic errors are hard to avoid in the background subtraction and normalization procedure. Since the total cross section does not vary over the temperature range of the measurements, this gives a constant temperature-independent error. The absolute cross sections are nevertheless fairly close to those obtained more reliably from the TOF measurements. In comparing data from the two spectrometers it should be noted also that the Q resolutions employed in the two cases are different so that the results obtained for comparable (mean values of) Q are somewhat different in magnitude as a result of the skewing of the scattering cross section towards low Q.

Results in Fig. 1(a) show the elastic scattering

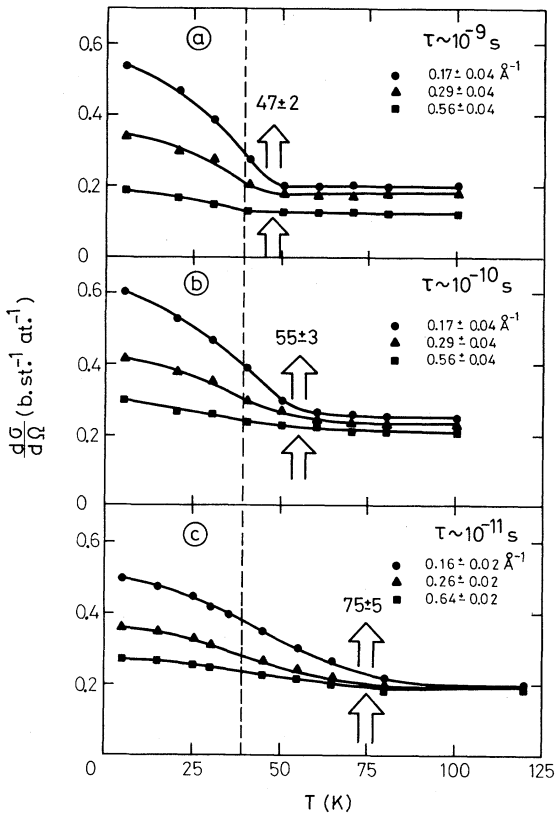

FIG. 1. "Elastic" scattering cross section vs temperature for several Q values: (a) Results from IN10 measurements with elastic energy resolution $\Delta E \sim 1.5$ μeV. (b) Total intensity within an energy window $\Delta E \sim 25$ μeV centered about $\omega = 0$ in the IN10 measurements. (c) Results from the IN5 time-of-flight measurements with elastic energy resolution $\Delta E \sim 230$ μeV. The arrows indicate the temperatures where marked increases in the cross sections with decreasing temperature begin. The dashed line marks the temperature of the ac susceptibility peak, measured with a time constant $\tau \sim 10^{-2}$ sec. The statistical errors are smaller than the size of the data points and the curves are drawn to guide the eye.

cross section for the Cu–8-at.%-Mn alloy obtained by fitting the spectra from the backscattering spectrometer IN10 to the measured elastic resolution function of the instrument [~ 1.5 μeV, full width at half maximum (FWHM)]. For a given Q the cross section is approximately temperature independent at high temperatures but begins to increase markedly below about 47± 2 K. The IN10 spectra span an energy range of ± 12 μeV about zero energy transfer. The total scattering within this energy range is shown plotted as a function of T in Fig. 1(b). It is seen that the marked increase with decreasing temperature in the latter case begins around a significantly higher mean temperature of 55± 3 K. The temperatures of the marked increases are determined graphically from the data and correspond to the temperatures where the rate of increase of the cross section has a maximum. The uncertainties in their determination are due to a statistical errors as well as to the fact that the temperature steps of the measured data points are fairly large.

The results for the elastic scattering cross section obtained using the IN5 TOF spectrometer are shown in Fig. 1(c). The TOF spectra show a broad quasielastic line whose energy width ranges from a minimum of ~ 1 meV (FWHM) to ~ 5 meV at high temperatures, and a central elastic line of width ~ 230 μeV.[7] The separation between the two was performed by fitting the measured quasielastic scattering to a Lorentzian function in the wings and interpolating in the central region. As in the previous cases, the elastic scattering cross section is approximately temperature independent at high temperatures but increases markedly below about 75± 5 K, a temperature which is significantly higher than in the cases where the energy resolution was 1.5 μeV and "25 μeV," respectively. As mentioned earlier the high-temperature scattering represents purely the nuclear incoherent (nonmagnetic) contribution which in the TOF results in Fig. 1(c) is seen to be almost Q independent within experimental error, confirming the good random substitutional character of the alloy. Its Q dependence over a similar Q range in the IN10 data [Figs. 1(a) and 1(b)] is due to unavoidable systematic errors in the flat background correction and vanadium normalization.

The magnetic scattering $S_e(Q)$, i.e., the temperature-dependent part of the scattering cross section, is strongly Q dependent because of short-range magnetic correlations which are invariably present in all moderately concentrated spin-glasses. However, within each set of measurements the temperature dependence of $S_e(Q)$ is closely similar over the Q range investigated. Hence, each of the $S_e(Q)$-vs-T curves may be taken as representative of the temperature variation of the "EA order parameter" measured with finite time constants, and the mean temperature indicated by the arrows in Fig. 1 around which the marked increase of the elastic scattering cross sections occur may be taken as the freezing temperatures corresponding to the various time constants of the measuring probe. In the diagram, Fig. 1, we have included also a vertical dashed line marking the temperature of the maximum in the ac susceptibility of the same alloy measured with a time constant of $\sim 10^{-2}$ sec.[7] The results clearly show the systematic variation of the freezing temperature with the measurement time constant. As mentioned earlier, the Mössbauer effect provides another probe for local magnetization and the EA order parameter. Mn nuclei are, however, not suitable for the study and so indirect methods using small amounts of Fe and Sn as probes in *Cu*-Mn alloys have been used.[8] These invariably give higher transition temperatures compared with those obtained from susceptibility measurements,[8] in good agreement with the above trend.

In conclusion, the present results give clear evidence for a wide spectrum of relaxation times of the spin system at temperatures around and below the "freezing temperature" and demonstrate the strong influence of the finite time constant of a probe on the result of measurement of a physical quantity such as the order parameter. Although it is possible to delineate a freezing temperature from each of the "order parameter" versus temperature curves, it is clearly not appropriate to regard them as sharp critical temperatures, as is often mistakenly done for the Mössbauer results. It is not unreasonable to expect that even the temperature of the ac susceptibility peak would shift to somewhat lower values if significantly longer measurement times could be used. Such a variation of the susceptibility maximum with the time constant of measurement between 10^{-11} and 10^{-2} sec has been shown earlier.[7] Overall, the results appear to suggest a saturation of the variation and so the question remains whether measurements on an infinite time scale could show the critical behavior predicted by the theories.

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High-Temperature Spin Dynamics of *Cu*-Mn Spin-Glasses

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Measurements of spin dynamics of *Cu*-Mn alloys containing between 1 and 7 at.% Mn reveal the phenomenon of exchange narrowing in neutron scattering, the Korringa relaxation mechanism being responsible for the intrinsic broadening of the paramagnetic spectrum, which is well described by a simple Lorentzian in the dilute limit. Additionally, the spin-spin exchange leads to enhance broadening of the spectrum at higher q's and generally to departures from the simple Lorentzian form of the spectral function.

Neutron scattering measurements on many paramagnets show broad quasielastic spectra at finite wave vectors due principally to the exchange interactions among the spins.[1] In dilute alloys such as *Cu*-Mn the broadening of the paramagnetic scattering may result not only from exchange interactions between the spins but is also expected to be due to the Korringa relaxation of isolated spins mediated via their exchange coupling with the conduction electrons which are in turn coupled to the lattice.[2]

Below I report a detailed neutron-scattering investigation of the dynamics of spins at room temperature in a series of *Cu*-Mn alloys containing between 1 and 7 at.% Mn which show clearly the contributions of the two processes, i.e., the Korringa mechanism and the spin-spin exchange coupling to the paramagnetic spectral function of the spins.

The paramagnetic neutron-scattering cross section for a system of N spins may be written as[3]

$$\frac{d^2\sigma}{d\Omega\, d\omega} = N\left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k_0} S(q,\omega), \qquad (1)$$

where

$$S(q,\omega) = 2\left\{\frac{\omega}{1-\exp(-\omega/k_B T)}\right\} F^2(q)\frac{\chi(q)}{g^2\mu_B{}^2} f(q,\omega). \qquad (2)$$

The expression inside the curly brackets is the detailed-balance factor, $F(q)$ is the magnetic form factor of the localized spins, $\chi(q)$ is the static susceptibility, and $f(q,\omega)$ is the spectral function representing the dynamics of the spin system.

It may be remarked that $\chi(q)$ is q independent for a noninteracting spin system and $F(q) \simeq 1$ for low q's. However, for the moderately concentrated spin-glasses studied here, $S(q,\omega)$ is a strongly varying function of q so that a proper account of its q dependence needs to be taken in the analysis of the time-of-flight spectra where the momentum transfer q varies with ω (for fixed scattering angles θ).

The measurements were made on the IN5 time-of-flight spectrometer at Institut Laue-Langevin using neutrons of incident wavelength 4.8 Å. The