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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

overall elastic instrumental resolution was 230 μeV. The data were corrected for background scattering and self-shielding of the sample and normalized with respect to vanadium. Phonon contribution was corrected for by subtracting off the measured scattering from a pure Cu sample of identical dimensions. The data points thus obtained were then put into constant-q format using a cubic spline interpolation routine. One disadvantage here is that the energy range spanned by a given constant-q cut through the closely spaced mesh of points in the q-ω plane decreases rapidly at low q's such that for $q < 0.4$ Å^{-1} it becomes too restricted to enable reliable fits to the spectral functions which have comparable half-widths at this temperature.

Another method of analysis which yields equivalent results over a wider ω range is the following. In practice, for the alloys studied a Lorentzian gives a reasonable fit to the bulk of the spectral profile $S(\theta,\omega)$ I use it to obtain the integral $S(q_\theta) = \int S(\theta,\omega)\,d\omega$, where q_θ are the elastic scattering vectors for each θ. Using the resultant set of values of $S(q_\theta)$ vs q_θ we then obtain by interpolation the correction factor $S(q_\theta)/S(q)$ for each of the points $S(\theta,\omega)$ (whose q value is defined by θ and ω). This factor attempts to reflect the data points into constant q_θ planes. I then recompute the integral $\int S(\theta,\omega)[S(q_\theta)/S(q)]d\omega$ to obtain a new set of values $S(q_\theta)$ and repeat the process. After a few iterations we obtain the final set of values of $S(q_\theta)$ such that $S(\theta,\omega)S(q_\theta)/S(q) \cong S(q_\theta,\omega)$. The constant-$q_\theta$ results obtained by this iterative procedure are in very good agreement with those obtained by the constant-q interpolation technique, in the energy range where direct comparison is possible, and give confidence in the analysis.

I have attempted to fit the data analyzed by the above methods with a Lorentzian spectral function, $f(q,\omega)$. The fits obtained were remarkably good over the whole q range ($0.08 < q < 1.2$ Å^{-1}) for the 1-at.%-Mn alloy and also for the 3-at.%-Mn sample, Figs. 1 and 2, but became progressively worse at higher concentrations where, at low q's, additional intensity in the high-energy wings was left over in the fits. In earlier measurements on a Cu–8-at.%-Mn alloy[4] the energy range ω of the data analyzed using the constant-q interpolation routine was too narrow to show deviations from a simple Lorentzian spectral function in the high-energy wings especially at low q's. I should mention that calculations of multiple-scattering contribution, under certain simplifying assumptions, yield correction factors of the order of 3% to 5% (varying with energy transfer) for the higher-concentration alloys. The distortion of the spectral shape normally resulting from these multiple-scattering effects is, however, too small to account for the large high-energy wings of the spectra in these higher-concentration samples.

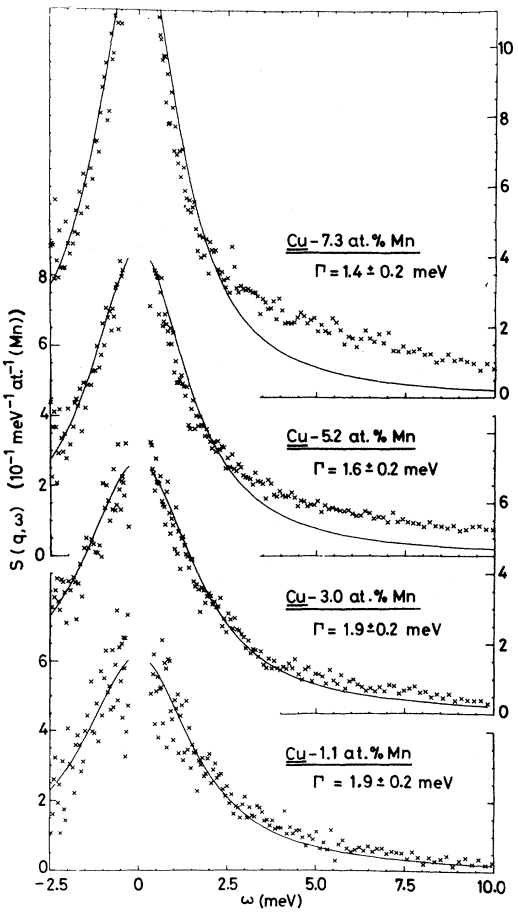

FIG. 1. $S(q,\omega)$, in units of meV^{-1} per Mn atom, vs ω for $q = 0.08$ Å^{-1} at $T = 300$ K, for Cu-Mn alloys with Mn concentrations as indicated in the diagram. The data points for elastic scattering are not shown in the diagram. The continuous curves represent the best fits to the data using the Lorentzian form for the spectral function $f(q,\omega)$. Note the progressive narrowing of the spectra accompanied by increasing deviation from the fits in the high-energy wings with increasing concentration.

In Fig. 3 I show the q variation of the half-widths Γ obtained from the Lorentzian fits to the spectra for the series of alloys. Note that the q dependence of the spectral widths gets stronger with increasing concentration and extrapolation to $q = 0$ yields constant values Γ_0 which decrease

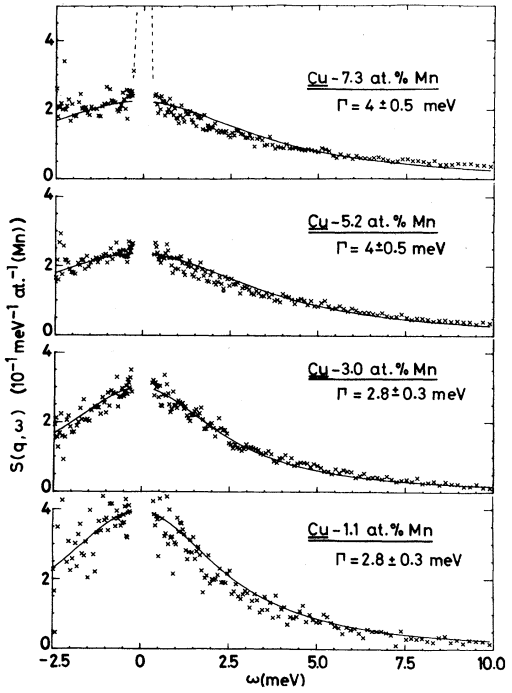

FIG. 2. $S(q,\omega)$ vs ω for $q = 0.9$ Å^{-1} at 300 K for *Cu*-Mn alloys including the Lorentzian fits to the spectra (continuous curves) as in Fig. 1. The dashed line in the uppermost part of the diagram shows the approximate form of the elastic scattering.

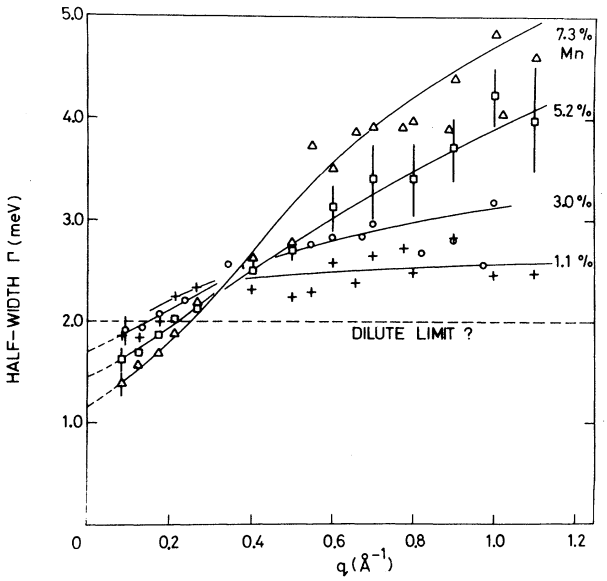

FIG. 3. The q dependence of the half-width Γ for the *Cu*-Mn alloys at 300 K. Error bars are only shown for one of the samples and are typical of the alloys, where the scatter among the data points indicates the uncertainly of the fits. The curves are drawn to indicate the trend of the data points.

with increasing concentration. Since increasing concentration implies increasing exchange interactions among the spins, the observed behavior suggests exchange narrowing which so far has only been observed in nuclear and electron spin resonance measurements. The shape of the spectral functions at low q's, Fig. 1, is also consistent with the exchange narrowing mechanism which requires that the fourth moment $\langle\omega^4\rangle$ is increased while the second moment $\langle\omega^2\rangle$ remains unchanged. This is achieved by narrowing of the line in the center and shift of some of the intensity into the wings, as observed.

The moments of the spectral function for a Heisenberg paramagnet have been calculated by de Gennes.[5] His formula for the second moment is

$$\langle\omega^2\rangle = \tfrac{8}{3}S(S+1)\textstyle\sum_j J_{ij}{}^2(1-\sin q\gamma_{ij}/q\gamma_{ij}), \qquad (3)$$

where J_{ij} is the exchange interaction between spins i and j at a distance r_{ij}. At low q's we obtain $\langle\omega^2\rangle \propto q^2$ so that Heisenberg exchange gives no contribution to the second moment as $q \to 0$. Also it accounts for the initial q^2-like increase of the linewidth (above the constant levels Γ_0).

At large q's the formula, Eq. (3), gives the asymptotic value $\langle\omega^2\rangle = \frac{8}{3}S(S+1)\sum_j J_{ij}{}^2$. Again, since $J_{ij}{}^2$ is proportional to the concentration, the approximately linear increase of the linewidth Γ with concentration at large q's suggests qualitative agreement with the formula.

The broad quasielastic linewidth Γ_0 as $q \to 0$ must clearly be due to the Korringa coupling of the localized Mn spins with the conduction electrons.[2] This should give rise to q-independent linewidths as shown by the dashed horizontal line in Fig. 3 which gives the expected behavior in the dilute limit. Lack of theoretical models, however, prevents a quantitative analysis of the shape of the spectral function especially since the two relaxation channels are not expected to contribute independently. It should be mentioned, however, that results of some earlier measurements on *Au*-Fe alloys[6] have been interpreted simply as a sum of two Lorentzians of different widths attributed to the Korringa relaxation of single spins (a broad Lorentzian of half-width $\Gamma \sim 10$ meV at 300 K) and of magnetic clusters (narrow line of half-width $\Gamma \sim 2$ meV at 300 K). However, the present results show that in the dilute limit the spectral function for Mn in Cu is a simple Lorentzian of relatively narrow half-width Γ

~2 meV at 300 K and that exchange interactions among the spins in the higher-concentration alloys leads to enhanced broadening at high q's accompanied by distortion of the spectral shape from a simple Lorentzian form, this distortion being particularly noticeable at low q's where with increasing concentration the spectral function appears progressively narrower in the central region with a shift of some of the intensity into the high-energy wings (exchange narrowing).

The dilute-limit Korringa width of ~2 meV at 300 K corresponds to a relaxation time constant of $\sim 10^{-13}$ s which is in reasonable agreement with the value of $\sim 10^{-11}/T$ s derived from the ^{63}Cu NMR measurements in dilute *Cu*-Mn alloys.[7] Using the relationship

$$\Gamma = \pi[J\rho(E_F)]^2 k_B T , \quad (4)$$

I obtain $[J\rho(E_F)] \sim 0.15$. It should be remarked that the ESR linewidth of Mn in Cu is extremely narrow by comparison, being about three orders of magnitude smaller than expected because of the well-known bottleneck effect.[8]

In conclusion, the present measurements of the dynamics of spin-glass alloys have permitted the following important observations. The paramagnetic scattering from the alloys has broad intrinsic width due to the Korringa relaxation of spins. The Heisenberg-type (Ruderman-Kittel-Kasuya-Yosida) exchange couplings between the spins then leads to "narrowing" of the lines at low q's and to increased broadening at higher q's. In the dilute limit the spectral function is closely represented by a Lorentzian over the q range investigated ($q \lesssim 1.2$ Å^{-1}) but gives only a crude description of the scattering from higher-concentration samples.

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