and broaden it. However, it does not seem likely from the 4.2 to 1.6 K trend (Fig. 2) that the residual increment in ΔH_0 as $T \to 0$ from 1.6 K will make a significant change. We feel that the discrepancy in ΔH_0 is real.

Most puzzling of all is the drastically different behavior observed by MA with *Cu*Mn alloys of lower concentration, but at comparable values of T/T_c to that found here at 4.2 K. It is especially strange that static line-broadening effects were absent in the MA results, in view of the fact that remanent (i.e., "static") magnetization phenomena have been reported for *Cu*Mn in the same concentration and temperature range.[10] The well-behaved scaling of remanent magnetization with concentration[10] would also appear to rule out the possibility of residual high-temperature Kondo phenomena in these systems.

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[3]S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).

[4]V. Cannella and J. A. Mydosh, in *Magnetism and Magnetic Materials—1973*, AIP Conference Proceedings No. 18, edited by C. D. Graham, Jr., and J. J. Rhyne (American Institute of Physics, New York, 1974), p. 651.

[5]A compilation of W values is given (Table I) by R. E. Walstedt and L. R. Walker, Phys. Rev. B 11, 3280 (1975).

[6]This assumption is better than it might appear at first glance. For example, the signs of $\vec{S}_i \cdot \vec{H}_0$ and $\Delta\vec{S}_i \cdot \vec{H}_0$ are clearly independent. The magnitudes of these quantities appear to be inversely correlated, but that effect is screened to some extent by the summations in Eq. (1).

[7]R. E. Walstedt and L. R. Walker, Phys. Rev. B 9, 4857 (1974).

[8]F. W. Smith, Phys. Rev. B 14, 241 (1976).

[9]V. Cannella, in *Amorphous Magnetism*, edited by H. O. Hooper and A. M. deGraaf (Plenum Press, New York, 1973), p. 195. The estimate of χ_{molar} given in the text is obtained by scaling the concentration from 1.3% to 0.96% and extrapolating the χ-vs-T curve to 1.6 K.

[10]J. Souletie and R. Tournier, J. Phys. (Paris), Colloq. 32, C1-172 (1971).

Reply to "Comment on 'Host Nuclear Resonance in a Spin-Glass: *Cu*Mn'"*

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Cu NMR linewidths and intensities in the spin-glass state of Cu + (0.6–1.0)-at.% Mn indicate appreciable freezing of the Mn spin system just below the spin-glass transition. Evidence has been obtained for considerable inhomogeneity in the dynamical behavior of the Mn spins. Comparison with our earlier results shows that Mn spin correlation times increase drastically between 0.2- and 0.6-at.% Mn.

In a previous Letter[1] we reported host Cu NMR measurements in the paramagnetic and spin-glass states of *Cu*Mn alloys with Mn concentrations in the vicinity of 0.2 at.%. No large increase of NMR linewidth was observed down to about half the spin-glass transition temperature T_0. This result appeared to be in conflict with mean-field theories of the spin-glass transition,[2] and with the muon spin-precession experiments of Murnick, Fiory, and Kossler[3] in Cu + 0.7-at.% Mn. In the preceding Comment Levitt and Walstedt[4] (LW) present host NMR data obtained from a 1.0-at.% Mn specimen, which give evidence for spin freezing at temperatures $T \lesssim 0.4T_0$. A preliminary report of similar measurements by the present authors will appear shortly.[5]

LW conclude that at lower Mn concentrations rapid fluctuations of the Mn spin system narrow the NMR line for temperatures down to $\sim 0.5T_0$, whereas at higher concentrations and for $T \lesssim 0.4T_0$ the Mn spin motion slows sufficiently to permit the observation of a large, field-independent line-

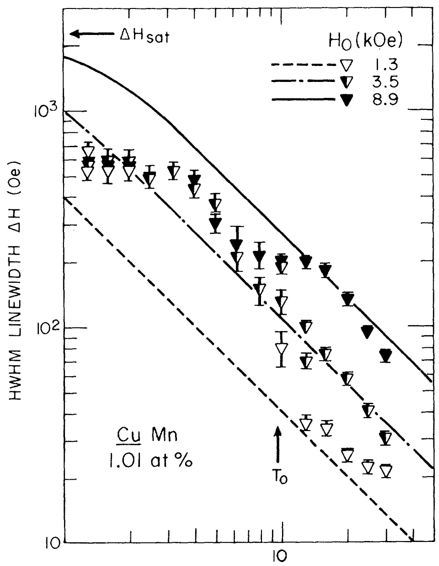

FIG. 1. Temperature and applied-field dependence of the Cu NMR linewidth (half-width at half-maximum) ΔH in Cu + 1.01-at.% Mn. The curves give Brillouin-function linewidths expected for independent Mn spins. Below $\sim 0.7T_0$, ΔH is practically field-independent, and in low applied fields has increased dramatically from its value above T_0. This behavior agrees with the muon-precession results of Ref. 3.

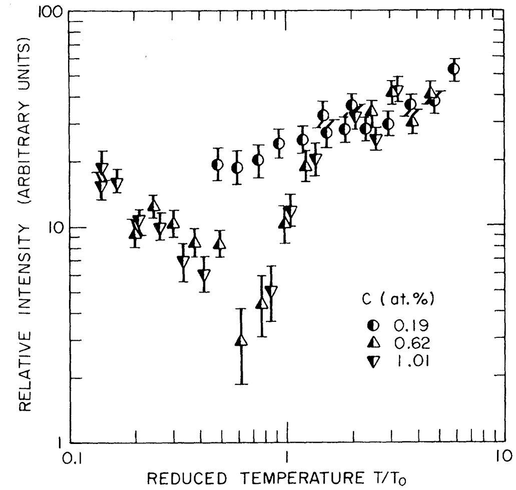

FIG. 2. Temperature and Mn-concentration dependence at an applied field of 3.5 kOe of the Cu NMR signal intensity in *Cu*Mn, relative to that expected from all nuclei in the specimen. The data have not been normalized. Measured intensities for the 0.62- and 1.01-at.% Mn specimens are similar functions of the reduced temperature T/T_0, and show a much more pronounced minimum at $\sim 0.6T_0$ than the intensity for the 0.19-at.% Mn specimen.

width. The purpose of this reply is to present experimental evidence that at higher concentrations [(0.6–1.0)-at.% Mn] spin freezing in fact sets in rather quickly below T_0, in a way which is not observed at lower concentrations[1,5] under comparable conditions of field and reduced temperature. Thus, Mn spin correlation times in the spin-glass state appear to increase drastically with Mn concentration between 0.2 and 0.6 at.%. We know of no explanation for this anomalous behavior.

We have observed Cu NMR in *Cu*Mn samples with nominal Mn concentrations of 0.19, 0.62, and 1.01 at.%, for applied fields H_a of 1.3, 3.5, and 8.9 kOe. Combined ^{63}Cu-^{65}Cu spin-echo spectra were recorded for three values of the time interval t between echo-forming pulses, to allow extrapolation of intensities and linewidths to $t = 0$. True linewidths ΔH were obtained by comparing the experimental spectra with calculated functions consisting of two overlapping Lorentzian lines of appropriate positions and intensities. Spin-glass transition temperatures T_0 were obtained from low-field ac susceptibility maxima measured in the same specimens. Figure 1 gives linewidth data obtained from the 1.01-at.% Mn specimen. For $H_a \gtrsim 3.5$ kOe, ΔH becomes sensibly independent of H_a below $\sim 0.7T_0$, and then tends toward the low-temperature value of about 600 Oe observed by LW. This is approximately half the value ΔH_r expected from a static random Mn spin configuration.[1,3] We recall that for Mn concentrations $\lesssim 0.2$ at.% a strong field dependence of ΔH was observed, and $\Delta H \ll \Delta H_r$ (for sufficiently low H_a) down to $\sim 0.5T_0$.[1]

Figure 2 gives the resonance intensity at 3.5 kOe (extrapolated to $t = 0$) relative to that expected from all nuclei in the specimen. A strong minimum is observed somewhat below T_0 for the 0.62- and 1.01-at.% Mn specimens, whereas for 0.19-at.% Mn the intensity varies much less rapidly with temperature. At 1.5 kOe this signal loss prohibits linewidth measurements for temperatures between 2 and 10 K (Fig. 1). The "lost" intensity is not recovered in an inhomogeneously broadened line of width $\sim \Delta H_r$, which would have been easily observable. We agree with LW that unobserved nuclei possess spin-echo decay times T_2 so short that the echo due to such nuclei decays before the spectrometer has recovered from rf pulse overload. Below $\sim 0.6T_0$, the intensity grows rapidly with decreasing temperature at the higher concentrations.

This raises the question of whether or not such short T_2 values are consistent with the "freezing" of the Mn spin system observed in the muon-precession experiments. The characteristic time which separates the rapid-fluctuation regime from the slow-fluctuation regime in resonance experiments is the spin precession period $(\gamma h_{loc})^{-1}$ in the fluctuating hyperfine field h_{loc}. Here γ is the gyromagnetic ratio of the spin probe (muon or Cu nucleus). In *Cu*Mn $(\gamma h_{loc})^{-1}$ fortuitiously takes on approximately the same value for muons as for Cu nuclei.[3,6,7] Thus, the transverse dynamical behavior of muons and Cu nuclei should be comparable. Muon experiments, with their time resolution of the order of nanoseconds,[3] sample combined static and dynamic broadening over a time scale short compared to $(\gamma h_{loc})^{-1} \sim 50/c$ nsec, where c is the Mn concentration in atomic percent. (This characteristic time is approximately the minimum value of both the free-precession decay time T_2^* and the spin-echo decay time T_2.) NMR spin-echo experiments, with rf pulse overload times of ~ 100 μsec, sample only those nuclei for which $T_2 \gtrsim 50$ $\mu\text{sec} \gg (\gamma h_{loc})^{-1}$. Thus, the unobserved nuclei in the Cu NMR experiments may possess T_2 values anywhere in the range 50 nsec to ~ 50 μsec for a 1-at.% Mn specimen. The upper limit of this range is consistent with either the rapid-fluctuation regime $1/T_2 = \gamma^2 h_{loc}^2 \tau_c$, or the slow-fluctuation regime $T_2 \sim \tau_c$, where τ_c is the Mn spin correlation time. This leads to 10^{-10} sec $< \tau_c < 10^{-5}$ sec. The fast-fluctuation regime, however, is not consistent with the muon results, for which the observed $T_2^* \sim (\gamma h_{loc})^{-1}$ indicates that $\tau_c \gtrsim T_2^*$. We conclude that for 1-at.% Mn, 50 nsec is a lower bound on τ_c for the slowest component of hyperfine-field fluctuation at the majority of spin-probe sites.[8]

For 1-at.% Mn, the static NMR linewidth of the observed Cu nuclei behaves similarly to that of the muons, for comparable applied fields.[9] This indicates that the distribution of the static component of hyperfine field is uniform to within a factor of 2 or 3 throughout the specimen. On the other hand, the appreciable loss of NMR intensity signals the presence of important inhomogeneity in the dynamical behavior of the Mn spins. This feature seems to us to be consistent with a "cluster" model of the spin-glass transition,[10] in which clusters are formed at temperatures somewhat greater than T_0, and below T_0 reorient with a wide distribution of characteristic times.

Our new data, as well as those of LW, differ qualitatively from the results reported previously.[1] The absence of large NMR linewidths below T_0 for low Mn concentrations is evidently caused by rapid Mn spin fluctuations ($\tau_c \sim 10^{-8}$ sec, $\gamma h_{loc}\tau_c \sim 10^{-2}$), but this rapid motion does not persist to higher concentrations. Extension of muon-precession experiments to lower concentrations, to obtain data on combined static and dynamic broadening with good time resolution, would be desirable; on the basis of our analysis, we would predict a long-lived component of muon precession ($T_2^* \sim 50$ μsec) below 0.2-at.% Mn. We are currently undertaking Cu NMR spin-lattice relaxation and spin-echo decay measurements over wide ranges of applied field, concentration, and temperature.

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[8]The upper bound on τ_c is not T_2, since the combination of a broad distribution of hyperfine-field components with $\tau_c \gg T_2$ and a rapidly fluctuating component $(\gamma h_{loc}\tau_c \ll 1)$ would also yield a broad line with the observed T_2.

[9]The increase of ΔH with decreasing temperature shown in Fig. 1 is more gradual than that observed in the muon experiments. This may be due to the correlation of the observed nuclei with regions of reduced hyperfine field as suggested by LW. Such an effect apparently does not decrease ΔH by as much as an order of magnitude.

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