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High-Frequency ac Susceptibility and ESR of a Spin-Glass

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We report the low-field ac susceptibility from 16 Hz to 2.8 MHz on the dilute spin-glass Ag:Mn in the vicinity of the glass temperature, T_G. Within experimental error T_G is frequency independent. We also report electron-spin-resonance measurements in the same samples at 1.6 and 9.1 GHz. The differences in the ESR data at two frequencies appear to be due to the different fields for resonance and not to the different measuring frequencies.

Several of the theories[1,2] which consider the zero-field–cooled spin-glass state[3] to be metastable predict an increase in T_G, the spin-glass temperature for the ac susceptibility maximum, with increasing ac measuring frequency, ν. All previously reported ac susceptibility measurements on spin-glasses have been made at relatively low frequencies (<5 kHz). However, electron spin resonance data taken at 9 GHz on spin-glass systems[4] have been characterized by the temperature at which the resonance g value departs from its high-temperature (paramagnetic) value, T_{gv}, or by the temperature where the resonance linewidth passes through a minimum value, T_{min}. Both these temperatures are considerably larger than the low-frequency ac-susceptibility T_G value and have been interpreted[5] as the effective glass temperature, $T_G{}^*$, at 9 GHz. In recent neutron-scattering experiments,[6] $T_G{}^*$ was taken as the temperature of the peak in the quasi-elastic-scattering cross section. Again $T_G{}^*$ was found to be greater than T_G. The effective measurement frequency was here assumed to be the reciprocal of the instrumental resolution time ($\sim 10^{-11}$ s). The large gap in frequency between previous ac measurements and the ESR and neutron measurements ($\sim 10^3$ to $\sim 10^{11}$ Hz) has prompted this work.

We report here new measurements on the low-field ac susceptibility, $\chi'(\nu,T)$, and ESR of dilute Ag:Mn spin-glass alloys in the vicinity of T_G. In particular our ac measurements extend to much higher frequencies (2.8 MHz) than in previous studies, and our ESR measurements are made at two well-separated frequencies. No evidence is found for any frequency dependence in the behavior of the ac-susceptibility spin-glass peak over five decades in frequency (16 Hz to 2.8 MHz). The quantitative differences seen in the ESR results in 1.6 and 9.1 GHz are shown to be due, at least in part, to the different fields for resonance and not to the different measuring frequencies.

Five master ingots of Ag:Mn with Mn concentrations of 1, 2, 3, 4, and 5×10^3 atomic ppm were made by arc-melting 5N (99.999% purity) Ag and 3N (99.9% purity) Mn. They were then given all the same homogenizing anneal at 800°C for 8 h *in vacuo*. All the measurements reported here were taken on samples made from the original five ingots. The ESR samples were grains produced by filing, having limiting dimensions, d, circa 300 μm. The low-frequency (LF) ac measurements (16 to 160 Hz) were made on the ESR samples. For the medium-frequency (MF) ac data (93 Hz to 109 kHz), such filings were sieved to produce a powder with $d \simeq 100$ μm. The high-frequency (HF) ac measurements (0.4 to 2.8 MHz) were made on 18-μm rolled foils. None of these samples were further annealed after the preparation treatment. The LF measurements were made with a commercial SQUID (superconducting quantum interference device) magnetometer, the MF measurements with a simple inductance bridge. The HF data were obtained by placing the sample in an inductor which formed part of the tank circuit of an oscillator. The oscillation frequency was then monitored as a function of sample temperature. In all the ac measurements the maximum amplitude of the alternating field was less than 2.5 G peak to peak. Conventional reflection techniques were employed for the ESR. In the MF ac and ESR experiments the samples were immersed in a pumped He^4 bath and their temperatures determined from the vapor pressure. In the low- and high-frequency ac measurements the samples were in an exchange gas and their temperatures measured using a germanium resistor. In these latter experiments, considerable care was taken to avoid thermal hysteresis and sample heating.

We exhibit in Fig. 1 representative plots at various frequencies of measured $\chi'(\nu,T)$ versus temperature for two 3000-ppm samples (LF and MF), and two 5000-ppm samples (MF and HF). We note the relative constancy of both shape and position within each family of curves (3000 LF, 5000 MF, etc.). The small differences in peak position between the 5000-ppm MF and HF samples are attributed to slight variations in mean concentration (circa 10%). The sharper nature of the HF peaks might be due to a smaller spread in concentration in the foil sample compared to the powder MF and grain LF samples. Considerable trouble was taken to produce samples with small limiting dimensions, d, because we found that, for d of the order of δ, the skin depth, the form of the $\chi'(\nu,T)$ curve becomes drastically modified. Briefly, if $d \simeq \delta/3$, there is an *apparent* monotonic decrease in χ' of the same magnitude as the real change in χ' for a 3000-ppm sample between 4 and 1 K. As d becomes greater than δ, the effect becomes much larger. The difference in shape of the 2.8-MHz curve in Fig. 1 with respect to those for 0.41 and 0.98 MHz is tentatively attributed to the onset of this skin-effect problem.

To reduce the error caused by noise and drift, T_G was taken at the maximum value of $\chi'(\nu,T)$ at any given frequency and a mean taken over several (3 to 9) temperature runs. We plot in Fig. 2 the mean spin-glass temperatures thus obtained versus $\log\nu$. On considering Figs. 1 and 2, we

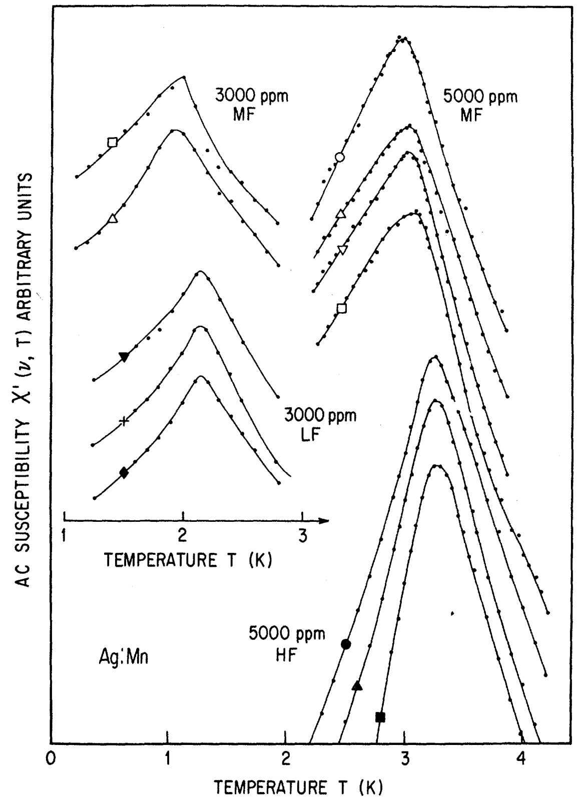

FIG. 1. Representative plots of $\chi'(\nu,T)$ vs temperature at various frequencies for some Ag:Mn samples. 3000 ppm LF: ♦, 16 Hz; +, 32 Hz; ▼, 160 Hz. 3000 and 5000 ppm MF: ○, 293 Hz; △, 2.9 kHz; ▽, 29 kHz; □, 109 kHz. 5000 ppm HF: ●, 0.41 MHz; ▲, 0.98 MHz; ■, 2.8 MHz. The types of samples and measurement techniques for the LF, MF, and HF ranges are given in the text. Each curve has been shifted arbitrarily on the vertical axis. The absolute values of the changes in susceptibility are constant to ±20% with change in frequency.

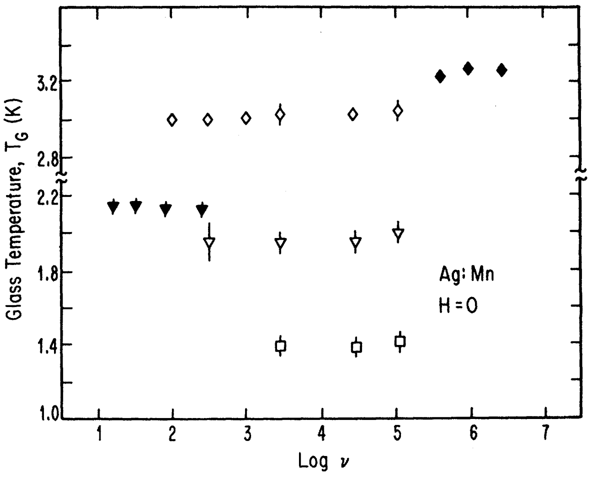

FIG. 2. The mean ac spin-glass temperature, T_G, vs the logarithm of the measuring frequency for different concentration Ag:Mn samples. ♦, 5000 ppm HF; ◇, 5000 ppm MF; ▽, 3000 ppm MF; ▼, 3000 ppm LF; □, 2000 ppm MF. The vertical error bars are equal to 2 standard deviations of the T_G measurements at a particular frequency.

conclude that within our experimental error, T_G for these samples is essentially frequency independent, certainly from 93 Hz to 109 kHz, and we feel this conclusion can be justifiably extended to cover the range 16 Hz to 2.8 MHz. Taking our limits of error in Fig. 2, the maximum admissible positive value for $\Delta T_G/T_G$ is 3% over at least three, and probably five, decades in frequency.

All our ESR measurements exhibit evidence for a linewidth minimum and a temperature-dependent g value. In Fig. 3 we show the values obtained[7] for T_{gv} at 1.6 and 9.1 GHz. The values of T_{gv} are lower at 1.6 GHz than at 9.1 GHz for each concentration. Both of these values are, however, larger than T_G measured at 29 kHz (also plotted in Fig. 3), as expected. Since the form of $\chi'(\nu, T)$ is strongly dependent on external field,[8] we repeated some of the ac measurements at 29 kHz in external magnetic fields of 600 and 3000 G. These fields, the ESR resonance fields at 1.6 and 9.1 GHz, respectively, were applied perpendicular to the ac measuring field, this being the same geometry employed for ESR. From these data we extracted a temperature T_B at which $\chi'(\nu, T)_{H\neq 0}$ "breaks away" from a Curie-like behavior. These values are also plotted in Fig. 3. We note that there appears to be a reasonable correlation between T_{gv} and T_B. In retrospect, this is not surprising. The departure from a Curie-law behavior for $\chi'(\nu, T)$ implies the presence of a static local field which would be expected to produce a shift in the field for magnetic resonance.

We show in Fig. 4 our measured ESR linewidth, ΔH, versus the reduced temperature, $T/c^{3/4}$. In all cases, the linear temperature dependence for ΔH breaks down, with a low-temperature increase beginning well above T_G. There are several features of these data we should like to note. In contrast to T_{gv}, the temperature for the minimum linewidth, $T_{\min}$, appears to be roughly the same for the two different ESR frequencies. The errors on $T_{\min}$ are rather large; if anything, $T_{\min}$(9.1 GHz)<$T_{\min}$(1.6 GHz). Hence, the use of $T_{\min}$ to imply a frequency-dependent glass temperature does not seem warranted. One cannot reasonably expect the characteristic temperatures plotted in Fig. 2 to double suddenly between 3 MHz and 1.6 GHz and then again be constant, or decrease slightly on going to 9.1 GHz. The value of $T_{\min}$ appears to obey the same scaling law as T_G (i.e., as $c^{3/4}$); in the inset to Fig. 4 we plot our values for T_G and $T_{\min}$, together with previous higher-concentration values. We note the good consistency of our low-concentration measurements with the earlier work, and that over a wide concentration range, $T_{\min} \simeq 2T_G$. Turning back to the main figure, we see that the overall behavior of the increasing contribution to H also scales as

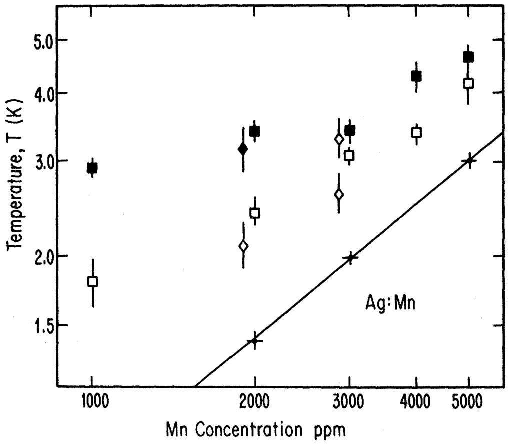

FIG. 3. T_{gv}, T_B, and T_G (29 kHz) vs concentration for dilute Ag:Mn. □, T_{gv}, 1.6 GHz; ■, T_{gv}, 9.1 GHz; ◇, T_B, 600 G; ◆, T_B, 3 kG; +, T_G, 29 kHz. For clarity, the values of T_B have been shifted to the left of their respective concentrations.

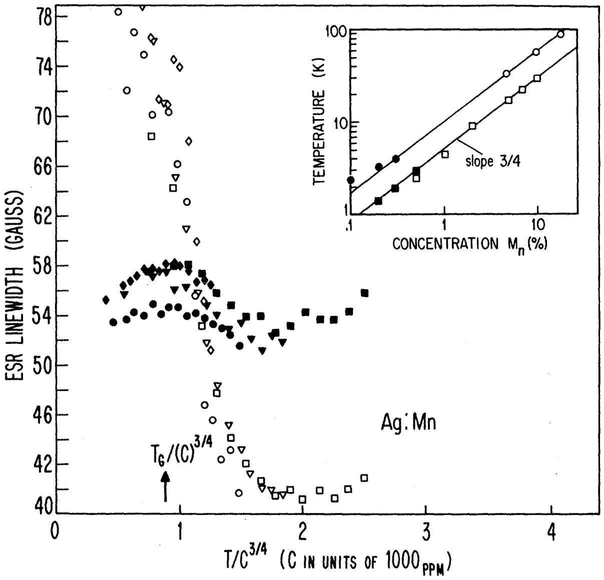

FIG. 4. ESR linewidth vs reduced temperature for Ag:Mn coarse-grain samples. 9.1-GHz measurements: ■, 2000 ppm; ▼, 3000 ppm; ●, 4000 ppm; ◆, 5000 ppm. 1.6-GHz measurements: corresponding open symbols. Inset: $T_{\min}$ and T_G vs Mn concentration for Ag:Mn: ●, $T_{\min}$, this work; ○, $T_{\min}$, Elliston (Ref. 4); ■, T_G (29 kHz), this work; □, T_G (circa 150 Hz), Ford and Mydosh (Ref. 7).

$c^{3/4}$ at each frequency. However, there is a striking contrast in the magnitude of this contribution at the different ESR frequencies (or fields). The exact source of this extra broadening (in any system[9]) is not altogether clear, and here the situation is probably complicated by the bottleneck in Ag:Mn. If one assumes the broadening to be due to local field fluctuations, our results imply that the larger external magnetic field for resonance at 9.1 GHz is quenching the fluctuation contribution to ΔH. This would also give a slight depression of $T_{\min}$ at 9.1 GHz with respect to 1.6 GHz, as perhaps observed.

In apparent direct contradiction to this work, there have been three very recent studies[10-12] reporting frequency-dependent spin-glass temperatures. However, two of these experiments were on rather high-concentration alloys.[10,11] Korn, Schilling, and Zibold[10] attribute their strongly frequency-dependent T_G in 23% Au:Co to metallurgical Co clusters behaving in accordance with the Nêel model.[1] Zibold[11] attributes the small shifts of T_G he observes in 8 and 12% Au:Fe to a frequency-dependent contribution to χ' from a small proportion of the Fe atoms which are in clusters. We thus conclude that there is no disagreement between these two experiments and our low-concentration results. There is, however, a clear discrepancy between our measurements and the work of Löhneysen, Tholence, and Tournier[12] on $LaAl_2$:Gd. These authors observe a 25% increase in T_G between 7.2 and 1142 Hz on an effective 2000-ppm sample. Apart from speculation on the differences between Mn- and Gd-based spin-glasses,[13] we are presently unable to explain the differences between our results and those of Ref. 12.

In conclusion, we have found the shape and position of the ac susceptibility peak in the dilute Ag:Mn spin-glass to be essentially independent of frequency over five decades. We propose that the differences in T_{gv} and $\Delta H(T)$ at the two ESR frequencies are functions of the applied magnetic fields alone. Our results do not lend any support to spin-glass models which predict a frequency dependence of the spin-glass behavior. Rather, the frequency-independent T_G and the ESR linewidth behavior are suggestive of some sort of phase transition.[14]

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