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DIFFERENCES BETWEEN SPIN GLASSES AND FERROGLASSES: Pd–Fe–Si

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Near the multicritical point in the magnetic phase diagram, some alloys that appear to be simple spin glasses actually have an intermediate ferromagnetic-like state between the high-temperature paramagnetic and low-temperature spin-glass states. The temperature dependences of the imaginary component of a.c. susceptibility and d.c. magnetization are presented to illustrate the subtle experimental differences between spin glasses and these ferroglasses.

THE APPEARANCE of spin-glass characteristics at low temperatures after a ferromagnetic transition at a higher temperature has been observed in a number of random alloy systems with competing magnetic exchange interactions. Alloys exhibiting this type of behavior are often referred to as "re-entrant spin glasses", or here, as "ferroglasses". Precise determination of the boundary line for the ferroglass-to-spin-glass transitions in the magnetic phase diagram, a microscopic description of the evolution of spin-glass behavior with the collapse of ferromagnetic behavior, and the nature of the spin-glass state at low temperatures are therefore topics of interest. In this communication, some of the subtle differences observed experimentally between spin glasses and ferroglasses are presented. Some ferroglasses deceptively appear to be simple spin glasses based on a peak in the vector or real susceptibility vs temperature. However, it is shown that the ferromagnetic state can be distinguished by the temperature dependences of the imaginary component of a.c. susceptibility and d.c. magnetization. A precise determination of the details of the magnetic phase diagram near the multicritical point can thus be made.

Figure 1 shows the real χ' and imaginary χ'' components of a.c. susceptibility for amorphous $Pd_{80-x}Fe_xSi_{20}$, $x = 21.1$, in an a.c. field H of 796 A m^{-1} (10 Oe) rms at a frequency f of 100 Hz. The magnitude of the vector susceptibility would be computed as $|\chi| = |\chi' - i\chi''| = (\chi'^2 + \chi''^2)^{1/2}$. No corrections were made for demagnetization factor, which is small for the sample geometry used here. The accuracy of the temperature measurement is estimated to be within $\pm$ 0.1 K. The curves are typical for ferroglasses. Upon cooling, χ' has an inflection point and χ'' continuously increases from zero at the Curie temperature $T_C = 161$ K. There is no distinguishing feature in χ' to identify spin freezing at a lower temperature T_{fg}; however there is a rounded peak in χ'' centered at 47.4 K that may be used to define T_{fg} [1]. T_{fg} corresponds to the temperature of maximum overall dissipation and is found to be very sensitive to field and frequency, decreasing as expected as H increases or as f decreases. It is proposed here that these are characteristics of ferroglasses.

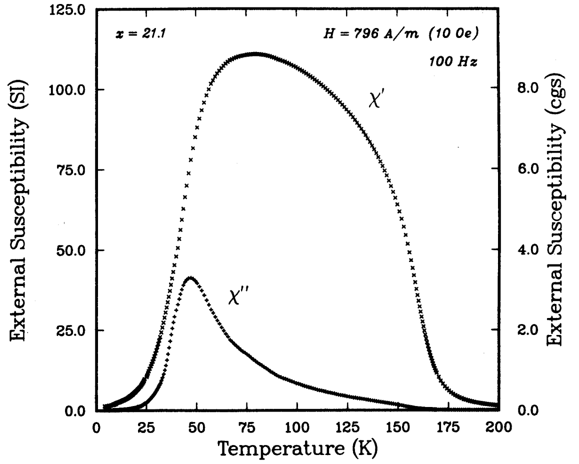

Fig. 1. Complex susceptibility for a ferroglass as a function of temperature.

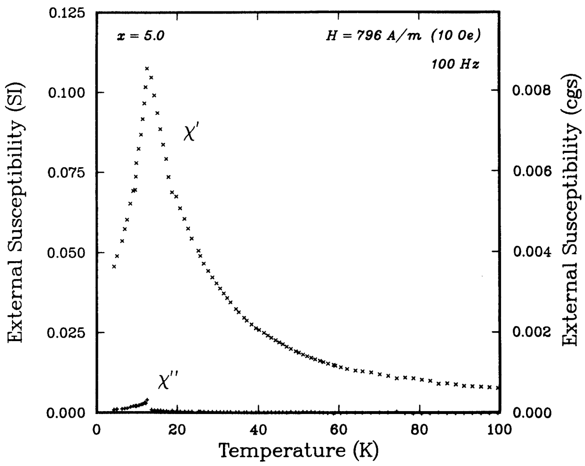

Fig. 2. Complex susceptibility for a spin glass as a function of temperature.

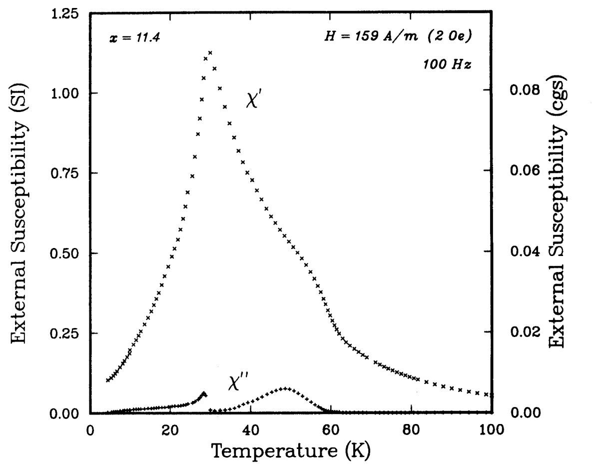

Fig. 4. Same as in Fig. 3, but for a smaller r.m.s. field.

Figure 2 shows χ' and χ'' for a simple spin glass, the $x = 5.0$ alloy. There is a sharp cusp in both χ' and χ'' at the same temperature, 12.6 K. χ'' is zero above the spin-freezing temperature T_f. T_f is found to be relatively insensitive to field and frequency. These are characteristics of true spin glasses in this alloy system.

The importance of the χ'' measurement in identifying T_{fg} in a ferroglass is illustrated in Figs. 3 and 4. Figure 3 shows χ' and χ'' for what appears to be a spin glass, the $x = 11.4$ alloy, but which, upon careful examination, turns out to be a ferroglass. The peak in χ' is rounded, not cusped. It has a maximum at $T_{max} = 29.9$ K, which is above T_{fg} as defined by the peak in χ'' at 27.9 K. χ'' is not zero above T_{fg}. T_{fg} is weakly field and frequency dependent. As further evidence that this alloy is actually a ferroglass, Fig. 4 shows the results in a smaller field of 159 A m^{-1} (2 Oe). T_{fg} is higher at 28.3 K, while T_{max} is about the same at 29.7 K. The Curie transition becomes apparent as a second peak in χ'' at 59 K. This feature is known to occur in other ferroglasses at T_C when measured in small fields.[2]

Another set of data for the $x = 11.4$ alloy, not shown, was taken in the usual field of 796 A m^{-1} (10 Oe) but at 10 Hz. The peak in χ'' occurs at $T_{fg} = 26.7$ K, the maximum in χ' is at $T_{max} = 29.1$ K, and T_C is 59 K. These low-frequency a.c. characteristic temperatures are compared with *d.c. measurements* in a field of 796 A m^{-1} (10 Oe) in Fig. 5. Making such a comparison is not unreasonable owing to the weak frequency dependence of T_{fg}. The field-cooled (FC) and

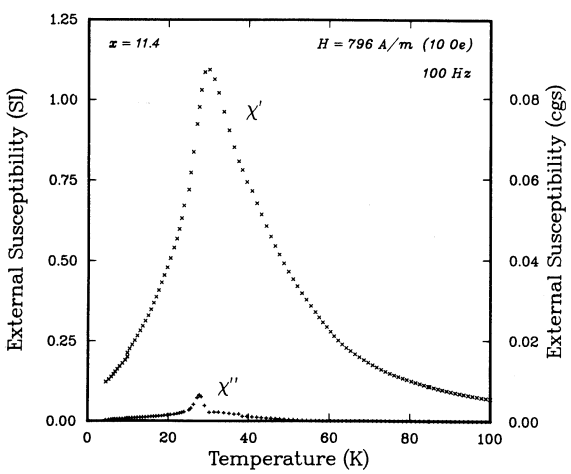

Fig. 3. Complex susceptibility for a ferroglass of composition close to the multicritical point in the phase diagram.

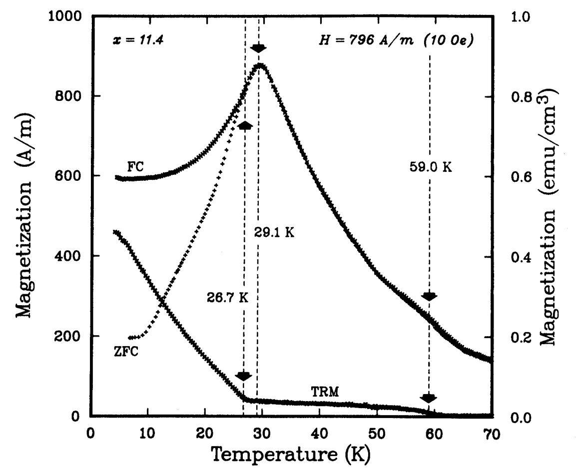

Fig. 5. Field-cooled (FC), zero-field-cooled (ZFC), and thermoremanent-magnetization (TRM) curves obtained in a d.c. field. The critical temperatures shown were obtained from a.c. susceptibility at a comparable r.m.s. field and low frequency. These temperatures are seen to coincide with features in the d.c. curves.

zero-field-cooled (ZFC) curves are shown. The thermoremanent-magnetization (TRM) curve was obtained by cooling in the field and measuring in zero field upon warming. The FC and ZFC curves merge and the TRM curve levels out very close to T_{fg} as determined from the 10-Hz measurement (26.7 K). The FC and ZFC curves both peak near T_{max} from the 10-Hz measurement (29.1 K). Finally, the TRM curve goes to zero and the FC and ZFC curves show slight bumps at T_C from the 10-Hz measurement (59 K).

It is concluded that the $x = 11.4$ alloy is a ferroglass, not a simple spin glass. Physically, this ferroglass region of the phase diagram is not characterized by long-range order; the external suceptibility in Figs. 3 and 4 is not limited by the reciprocal of the demagnetization factor. Rather, the existence of ferromagnetic correlation over a finite length is suggested. Experimental details of the measurements, a discussion of the peak in χ'' as a definition of T_{fg}, and the Pd–Fe–Si phase diagram will be presented in a forthcoming paper.

In summary, the main points are: (1) The low-field susceptibility peak is cusped in spin glasses, but rounded in ferroglasses. (2) Both the real (χ') and imaginary (χ'') components of susceptibility peak at the spin-freezing temperature T_f in spin glasses but at different temperatures in ferroglasses; the peak in χ'' defines T_{fg} in ferroglasses. (3) T_f is relatively insensitive to field and frequency in spin glasses while T_{fg} is more sensitive to these parameters in ferroglasses. (4) χ'' above T_f is zero in spin glasses, but may be greater than zero above T_{fg} (up to the Curie temperature T_C) in ferroglasses. (5) The thermoremanent magnetization (obtained by field cooling) decays, upon warming in zero field, to zero at T_f in spin glasses, but decays to a plateau value at T_{fg} (finally dropping to zero at T_C) in ferroglasses. (6) Field-cooled and zero-field-cooled magnetization curves merge at T_f as well as T_{fg}; this occurs at the temperature of maximum magnetization in spin glasses but below that temperature in ferroglasses. (7) Measurements of both the imaginary and real components of a.c. susceptibility provide a consistent way for determining the magnetic phase diagram, especially close to the multicritical point, for systems with competing interactions.

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