

## Frequency and field dependence of the ac susceptibility of the $AuMn$ spin-glass

C. A. M. Mulder, A. J. van Duyneveldt, and J. A. Mydosh  
*Kamerlingh Onnes Laboratorium der Rijks-Universiteit, Leiden, The Netherlands*  
 (Received 22 October 1981)

We have measured the ac susceptibility for  $AuMn$  containing 2.98 at. % Mn. Below the spin-glass freezing temperature  $T_f$  a small frequency dependence of  $\chi$  is found. The application of an external magnetic field near  $T_f$  shows a distinct reduction of  $\chi'$ . A comparison of the susceptibility behavior with that of other metallic spin-glass alloys containing manganese is made.

Over the past few years much interest has been devoted to the temperature, frequency, and magnetic field behavior of the differential (ac) susceptibility of spin-glasses. The cusplike behavior in the temperature dependence of the susceptibility remains one of the simplest and best methods for determining the freezing temperature  $T_f$ . More recently the frequency dependence has become a testing grounds for the various descriptions of the spin-glass transitions, e.g., equilibrium phase transition, metastable nonequilibrium state, or thermally activated processes.<sup>1</sup> Up until now at least a small frequency dependence of the susceptibility in the range 1 Hz to 1 kHz has been observed for every measured spin-glass system.<sup>2</sup> The field dependence represents a strong coupling into the frozen spin-glass state and greatly alters this state by introducing remanences, irreversibilities, and time dependences. Additional information concerning the spin-glass transition may be gained by studying the different field behaviors as measured by the ac susceptibility or, most recently with the use of SQUID techniques, by a dc susceptibility.<sup>2</sup> The present paper deals with an ac susceptibility investigation of the  $AuMn$  spin-glass in which the frequency and field dependences are determined at various temperatures.

The differential susceptibility is measured by means of the well-known mutual inductance technique (frequency range from 1 Hz to 10 kHz). Both the in-phase component  $\chi'$  and the out-of-phase component  $\chi''$  of the complex susceptibility are measured simultaneously. The absolute accuracy of determining  $\chi$  is better than 1%; however, the relative accuracy of  $\chi'$  at a fixed frequency is better than 0.1%. Measurements are performed in the temperature range from 1 to 30 K in the experimental setup described by Groenendijk *et al.*<sup>3</sup> The temperature could be kept constant to within 0.1%, while the absolute accuracy of the temperature determination is better than 0.5%.

An alloy of  $AuMn$  containing 2.98 at. % Mn was prepared by arc melting. The Mn concentration of the sample was determined by spectrophotometric analysis. Additionally the sample was annealed at

900 °C, followed by a rapid quench into ice water. In order to reduce as much as possible the effects of eddy currents and skindepth, the experiments were carried out on a finely powdered sample with a grain size that does not exceed 100  $\mu\text{m}$ . The sample did not receive any further heat treatment after the filing. Further, the  $AuMn$  powder was mixed with finely powdered, nonconducting  $Al_2O_3$  to avoid electrical contact between the  $AuMn$  grains. Without  $Al_2O_3$  the highest measuring frequency, where no artificial increase of the  $\chi''$  signal occurs, is about 200 Hz, while after mixing with  $Al_2O_3$  the averaged observed, low-frequency value of  $\chi''$  is found to be smaller by a factor of 2, and frequencies up to 2 kHz can be applied before the artificial  $\chi''$  results are obtained. The contribution of the diamagnetic  $Al_2O_3$  to the susceptibility of  $AuMn$  is negligibly small.

First, we measured the zero-field susceptibility as a function of the temperature. The result for  $\nu = 234$  Hz in the temperature range between 3 and 25 K is presented in Fig. 1.  $\chi'$  exhibits a sharp maximum which we identify with the freezing temperature,

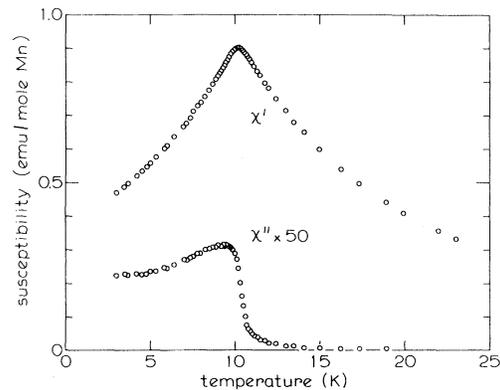


FIG. 1. Temperature dependence of the susceptibility  $\chi'$  and  $\chi''$  (applied oscillating field  $\leq 1$  Oe, measuring frequency 234 Hz) for  $AuMn$  (2.98 at. % Mn) in zero static external magnetic field.

$T_f(\nu=234 \text{ Hz}) = 10.23 \pm 0.05 \text{ K}$ . In addition, it is seen that a pronounced anomaly in  $\chi''$  is observable around this temperature, the maximum in  $|d\chi''(T)/dT|$  nicely coinciding with the maximum in  $\chi'(T)$ , i.e., with the freezing temperature  $T_f(\nu)$ . A similar behavior of  $\chi''$  around  $T_f$  was previously observed in  $\text{CuMn}$ ,<sup>4</sup>  $\text{PdMn}$ ,<sup>5</sup> and  $(\text{Fe}_{0.06}\text{Ni}_{0.94})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ .<sup>6</sup>

Second, the frequency dependence of  $\chi'$  around and below the freezing temperature was detected with an extremely high relative accuracy ( $\sim 0.004 \text{ K}$  at  $10 \text{ K}$ ). For temperatures around  $T_f$  the result for several frequencies is depicted in Fig. 2 on an expanded scale. We characterize the frequency dependence of the freezing temperature by the relative shift in  $T_f$  per decade of frequency, i.e.,

$$\frac{1}{T_f} \frac{\Delta T_f}{\Delta \log_{10} \nu} = (4.5 \pm 0.4) \times 10^{-3} .$$

This value is of the same order of magnitude as the one previously reported for  $\text{CuMn}$ ,  $(5.0 \pm 0.7) \times 10^{-3}$ ,<sup>7</sup> and  $\text{AgMn}$ ,  $(6 \pm 2) \times 10^{-3}$ ,<sup>8</sup> but substantially smaller than those for other diluted metallic spin-glasses containing Mn atoms, such as  $\text{PdMn}$  (0.011–0.015)<sup>5</sup> and  $\text{NiMn}$  (0.018).<sup>9</sup> For temperatures below  $T_f$  the various  $\chi'(\nu)$  curves remain separated, but slowly converge towards a single, nonzero, frequency-independent value at zero temperature. To illustrate this "converging" effect we have plotted the frequency dependence of  $\chi'$  (normalized to 234 Hz) for several temperatures in Fig. 3. This behavior is in accordance with the frequency dependence of  $\chi'$  in  $\text{CuMn}$ ,<sup>7</sup> but in contrast to the increase of the frequency dependence of  $\chi'$  as the tem-

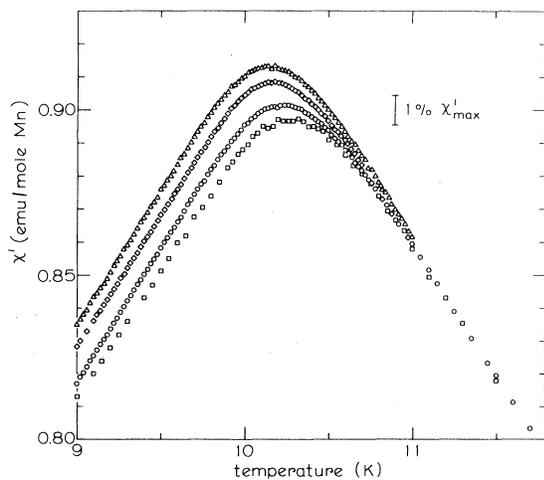


FIG. 2. Temperature dependence of  $\chi'$  around the freezing temperature  $T_f(\nu=234 \text{ Hz}) = 10.228 \pm 0.004 \text{ K}$  (absolute accuracy of the temperature determination 0.05 K) for  $\text{AuMn}$  (2.98 at. % Mn),  $\Delta$ : 3.7 Hz;  $\diamond$ : 29.3 Hz;  $\circ$ : 234 Hz;  $\square$ : 1.88 kHz.

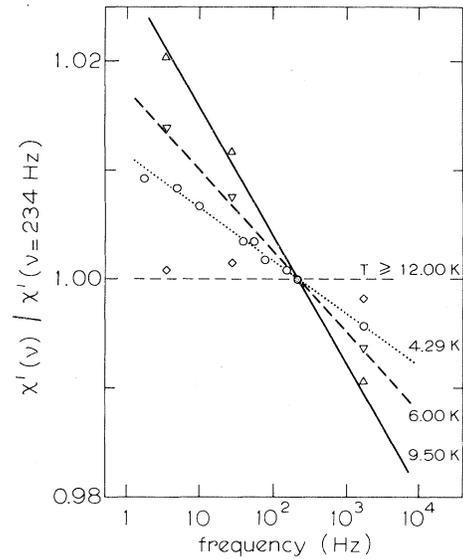


FIG. 3. Normalized susceptibility  $\chi'(\nu)/\chi'(\nu=234 \text{ Hz})$  as a function of frequency at several temperatures for  $\text{AuMn}$  (2.98 at. % Mn). The drawn lines are a guide to the eye only.

perature is lowered below  $T_f$  for other Mn-impurity alloys, such as  $\text{PdMn}$ <sup>5</sup> and  $\text{NiMn}$ .<sup>9</sup> A full comparison between the various different, frequency-dependent susceptibility measurements on Mn-containing, metallic alloys is planned to be treated in a subsequent paper.<sup>10</sup>

Finally, we examined the dependence of the susceptibility with respect to the external static magnetic field (up to 4.5 kOe) which is applied parallel to the ac driving field. In Fig. 4 we have plotted the field-dependent susceptibility as  $\chi'/\chi'_0$ , where  $\chi'_0$  is the zero-field susceptibility measured at the freezing temperature (measuring frequency 234 Hz). It should be

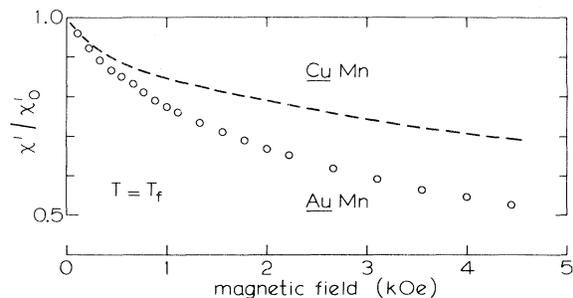


FIG. 4. Susceptibility  $\chi'$  divided by the zero-field susceptibility  $\chi'_0$  as a function of the static external magnetic field at  $T = T_f$  for  $\text{AuMn}$  (2.98 at. % Mn,  $T_f = 10.23 \text{ K}$ , circles). The  $\text{CuMn}$  (0.70 at. % Mn,  $T = T_f = 7.65 \text{ K}$ ) result from Ref. 7 is shown by the dashed line.

noted here that within the experimental accuracy the field dependences of  $\chi'$  are independent of the measuring frequency as long as  $T_f$  is properly scaled with the frequency. The dashed line represents similar measurements for quenched CuMn (0.7 at. % Mn,  $T_f = 7.65$  K),<sup>7</sup> which clearly show a much smaller dependence on the applied magnetic field. In addition, Fig. 5 presents the normalized change in the susceptibility  $(\chi'_0 - \chi)/\chi'_0$  versus the field at the freezing temperature  $T_f = 10.23$  K and at two temperatures, just above and just below  $T_f$ . It is seen that the field dependence of the susceptibility is strongest for  $T = T_f$ , while the suppression of  $\chi'$  with field becomes weaker at temperatures both above and below  $T_f$ . Note that these two latter curves show a rather similar field dependence with each other, indicating that  $T_f$  behaves as a symmetry point with respect to  $\chi'(H)$ . This is supported by the fact that we did not observe  $T_f$  to shift on applying an external magnetic field within the experimental accuracy. As was already pointed out in Ref. 7, the application of critical exponents and scaling laws to the field dependence proves to be of little validity. In fact, the slope for  $T = T_f$  in the log-log plot of Fig. 5 gradually decreases from 2 at 10 Oe towards  $\frac{1}{2}$  at 5 kOe.

A comparison of the ac susceptibility results with dc (or SQUID) magnetization measurements on CuMn, AgMn, or AuMn<sup>11-15</sup> is of interest. First the ac susceptibility is fully reversible in an external field applied parallel to the ac driving field. The influence of changing the frequency can be taken into account by scaling the freezing temperature. Then the field dependence of  $\chi'$  is exactly the same at the different measuring frequencies. In contrast the dc magnetization is only reversible and independent of time after field cooling, and then as long as the field is kept constant. Here one usually defines  $\chi_{\text{static}} = M/H$ . The field-cooled dc magnetization is a combination of  $M$  reversible and  $M$  irreversible (= thermal remanent magnetization in the field cooling field).<sup>15,16</sup> Therefore it is not possible to directly compare the ac- $\chi'$  and  $\chi_{\text{static}}$  without taking into account  $M$  irreversible and its field dependence. By field cooling a spin-glass one prepares the system in its infinite time state. Thus  $M$  irreversible is well defined (time independent), but one must measure this quantity as a function of the field cooling field and then subtract it

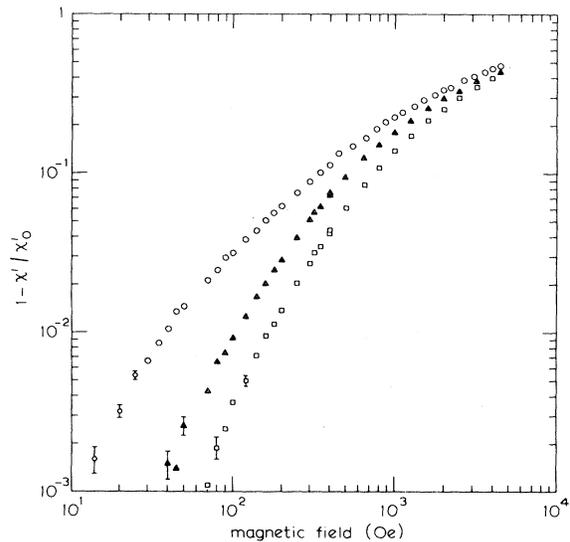


FIG. 5. Normalized change in susceptibility  $(\chi'_0 - \chi)/\chi'_0$  as a function of magnetic field at several temperatures for AuMn (2.98 at. % Mn);  $\circ T = T_f = 10.23$  K,  $\blacktriangle T = T_f + 1.0 = 11.2$  K,  $\square T = T_f - 1.0 = 9.2$  K.

from the total magnetization in order to obtain  $M$  reversible. Up until now such a comparison of  $M$  reversible with ac- $\chi'$  has not been performed. A full discussion of this point and the theoretical treatments of the various susceptibilities will be reviewed for a future publication.<sup>10</sup>

We conclude from our dynamical susceptibility measurements that the freezing temperature of AuMn is only weakly dependent on the frequency while below  $T_f$  the frequency of  $\chi'$  further diminishes, a result which is similar to the one observed for CuMn. The application of relatively small external magnetic fields strongly reduces and rounds the peaks in  $\chi'(T)$ .<sup>17</sup> The reduction with magnetic field for AuMn is somewhat stronger than that found for CuMn.

This work was supported in part by the Nederlandse Stichting voor Fundamenteel Onderzoek der Materie (FOM).

<sup>1</sup>For a recent survey of spin-glass theories see *Proceedings of the International Conference on Disordered Systems and Localization*, edited by C. Di Castro (Springer, Berlin, 1981).

<sup>2</sup>See the collection of spin-glass papers in *Physica (Utrecht) 107B* and *108B* (1981).

<sup>3</sup>H. A. Groenendijk, A. J. van Duynveldt, and R. D. Willett, *Physica (Utrecht) 101B*, 320 (1980).

<sup>4</sup>C. A. M. Mulder, Ph.D. thesis (University of Leiden, 1982) (unpublished).

<sup>5</sup>C. A. M. Mulder, A. J. van Duynveldt, H. W. M. van der Linden, B. H. Verbeek, J. C. M. van Dongen, G. J. Nieuwenhuys, and J. A. Mydosh, *Phys. Lett. A* **83**, 74 (1981).

<sup>6</sup>L. Lundgren, P. Svedlindh, and O. Beckman, *J. Magn.*

- Magn. Mater. 25, 33 (1981).
- <sup>7</sup>C. A. M. Mulder, A. J. van Duyneveldt, and J. A. Mydosh, Phys. Rev. B 23, 1384 (1981).
- <sup>8</sup>J. L. Tholence, Physica (Utrecht) 108B, 1287 (1981).
- <sup>9</sup>R. B. Goldfarb and C. E. Patton, Phys. Rev. B 24, 1360 (1981).
- <sup>10</sup>M. Hardiman, C. A. M. Mulder, and A. J. van Duyneveldt (unpublished).
- <sup>11</sup>S. Nagata, P. H. Keesom, and H. R. Harrison, Phys. Rev. B 19, 1633 (1979).
- <sup>12</sup>R. V. Chamberlin, M. Hardiman, and R. Orbach, J. Appl. Phys. 52, 1771 (1981).
- <sup>13</sup>P. Monod and H. Bouchiat, in *Proceedings of the International Conference on Disordered Systems and Localization*, edited by C. Di Castro (Springer, Berlin, 1981).
- <sup>14</sup>A. P. Malozemoff and Y. Imry, Phys. Rev. B 24, 489 (1981); B. Barbara, A. P. Malozemoff, and I. Imry, Physica (Utrecht) 108B, 1289 (1981).
- <sup>15</sup>S. T. McAlister and C. M. Hurd, J. Magn. Magn. Mater. 15-18, 169 (1980).
- <sup>16</sup>J. L. Tholence and R. Tournier, J. Phys. (Paris) 35, C4-229 (1974).
- <sup>17</sup>V. Cannella, in *Amorphous Magnetism*, edited by H. O. Hooper and A. M. de Graaf (Plenum, New York, 1973), pp. 195-206.