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Observation of ferromagnetic spin waves in a "reentrant" Ni-Mn alloy

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Neutron scattering measurements in a "reentrant" Ni-Mn alloy reveal well-defined spin waves at all temperatures below T_c (330 K), particularly and for the first time, below T_f (40 K) the temperature of the down turn in the ac susceptibility. The spin waves coexist with intense quasielastic scattering which develops below ~ 100 K, and the spin-wave damping, the stiffness constant, and the transverse susceptibility are all enhanced. The results contradict the common hypothesis of a breakup of the infinite ferromagnetic network in the reentrant state at T_f.

Magnetic systems with competing ferromagnetic (FM) and antiferromagnetic (AF) interactions such as $Eu_xSr_{1-x}S$ (Ref. 1), $Au_{1-x}Fe_x$ (Ref. 2), and $Pd_{1-x}Mn_x$ (Ref. 3) usually freeze into a spin-glass (SG) state when positive and negative interactions roughly balance each other. When FM bonds are predominant, long-range ferromagnetic order sets in at a well-defined temperature, T_c. Among the highly debated questions in the domain of spin-glasses is that of the behavior of these systems between these two limits close to the critical threshold for ferromagnetic long-range order. For intermediate composition, ac susceptibility (χ_{ac}) measurements exhibit a sharp increase at T_c followed by a plateau limited by demagnetizing effects down to a temperature T_f where the ac susceptibility decreases again.[3] While T_c is unambiguously ascribed to the onset of long-range order, T_f has been tentatively associated with a breakdown of the FM network and a transition to a spin-glass-like state.[3]

Such a "reentrant" behavior is indeed found in the Sherrington-Kirkpatrick model for Heisenberg spins.[4] The calculated phase diagram shows transitions from FM towards two mixed phases associating ferromagnetism and a SG freezing of the transverse components of the spins. Whether the real systems follow this pattern remains controversial. It should be noted that χ_{ac}, being strongly influenced by the onset of anisotropy and hysteresis behavior, cannot give unambiguous information.[5] However, the existence of a change in the magnetic state, when the temperature is further reduced below T_c, is strongly suggested by small-angle neutron scattering experiments which reveal in all these systems an anomalous increase of the scattering below a characteristic temperature T_f' higher than T_f.[6–8] The nature of the low-temperature state below T_f' remains unclear. Recent Mössbauer studies on $Au_{1-x}Fe_x$ (Refs. 9 and 10) shed some light as a progressive canting of the spins was evidenced below T_f'. No evidence for a sharp transition could be found.[9] Inelastic neutron scattering in the $Fe_{1-x}Cr_x$ system[11] revealed well-defined spin waves below T_c in the FM state. However, an anomalous decrease of their stiffness constant was observed on further reducing the temperature and no spin waves could be detected in the presence of the quasielastic peak. More recent results on the amorphous Fe-Ni alloys[12] show that the spin waves and the quasielastic peak coexist in a restricted range of temperature. However, at lower temperature, below the downturn of the ac susceptibility, the spin-wave spectrum is found to collapse.[12] These results are held as indicating a breakdown of the long-range ferromagnetic order at low temperatures.[11,12]

In the following, we present a closely similar inelastic scattering study of a $Ni_{78.4}Mn_{21.6}$ polycrystalline alloy. Ni-Mn alloys are good examples of competing interactions systems: Their magnetic phase diagram, derived from ac susceptibility data, is quite similar to those of the other reentrant systems. For Mn concentrations $C_{Mn}=27$–30 at.% they behave as normal spin-glasses exhibiting a peak in the ac susceptibility and usual remanence properties.[13] For $20 < C_{Mn} < 26$ at.% the χ_{ac} data show the typical plateau between T_c and T_f (Ref. 14): For the $Ni_{78.4}Mn_{21.6}$ studied here by neutron scattering $T_c = 330$ K while $T_f \simeq 40$ K (Ref. 15). In contrast to previous studies, we observed well-resolved spin waves down to the lowest temperature, in particular, well below T_f, which demonstrates that the long-range ferromagnetic order is preserved at all temperatures. Below $T \simeq 100$ K, the spin waves coexist with the usual characteristic quasielastic scattering. No decrease of the stiffness constant D is found when the spin waves are described by a damped harmonic oscillator (DHO) function, while a decrease of D is indeed found when using a double Lorentzian (DL) model.

Inelastic neutron scattering measurements were carried out on cold source three-axis spectrometers: 4 F at the reactor ORPHEE in Saclay and IN 12 at the high-flux reactor of the Institut Laue-Langevin in Grenoble. On IN 12 we used neutrons of incident wave vector 1.4 Å^{-1}. Inelastic scattering has been measured at scattering vectors $Q = 0.04$, 0.045, 0.055, 0.065, and 0.075 Å^{-1} between room temperature and 1.4 K. Careful attention has been paid to the subtraction of the background and nuclear scattering and to the calculation of the effective Q values.[16] A set of corrected data for $Q = 0.055$ Å^{-1} is presented in Fig. 1 in order to illustrate the temperature dependence of the scattering. Well-defined spin waves are observed in the whole range of temperature down to $T = 16$ K. When the temperature is lowered below 180 K, an apparent shift of the spin-wave energy towards

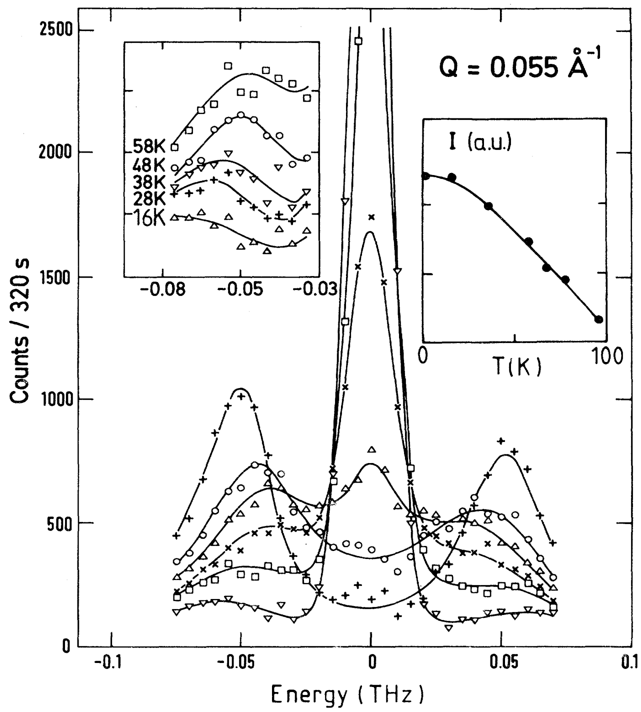

FIG. 1. Inelastic spectra for $Q = 0.055$ Å^{-1} at several temperatures: $T = 180$ K (+); 120 K (○); 100 K (△); 80 K (×); 58 K (□); 38 K (▽). The background and nuclear scattering have been subtracted. Spin waves are observed at all temperatures. Below about 100–120 K they coexist with a quasielastic scattering. The lines drawn through the data are calculated intensities using the damped harmonic oscillator model (see text). The left insert shows the detailed variation of the inelastic scattering in the energy range −0.08–0.03 THz at temperatures below 58 K. The right insert shows the temperature dependence of the integrated intensity I of the quasielastic scattering. The line drawn is a guide to the eye.

lower values is observed, associated with an increase of their linewidth. However, at lower temperatures, below about 70 K, the spin-wave energy is found to increase again. Below about 100–120 K a quasielastic scattering appears whose intensity increases rapidly as the temperature decreases. Spin waves coexist with the quasielastic signal at all temperatures below ~100 K.

The scattering cross section for magnetic coherent inelastic scattering from a FM can be expressed as[17]

$$S(Q,\omega) = A\frac{\omega}{1-\exp(-\hbar\omega/k_BT)}\chi_Q^t(\omega=0)F_Q^t(\omega) \quad . \tag{1}$$

In the limit $\hbar\omega << k_BT$ which is satisfied in our experiments, Eq. (1) reduces to

$$S(Q,\omega) \propto T\chi_Q^t(\omega=0)F_Q^t(\omega) \quad ,$$

where $\chi_Q^t(\omega=0)$ is the transverse part of the susceptibility response to a static sinusoidal field of wave vector Q, and $F_Q^t(\omega)$ is the spectral function which describes the dynamics of the system (dispersion and damping) and obeys the sum rule $\int_{-\infty}^{\infty} F_Q^t(\omega)d\omega = 1$. In the FM state we can then describe the scattering by using three parameters χ_Q^t, D, and Γ, where D is the stiffness constant of the spin waves which obey the dispersion law $\omega_Q = DQ^2$ and Γ is their intrinsic linewidth. Unfortunately, there is no definitive formulation for spectral function $F_Q^t(\omega)$. We have analyzed our data with the two commonly employed forms, namely, the double Lorentzian (DL),

$$F_{Q(\omega)}^t = \frac{1}{2}\pi\left[\frac{\Gamma}{(\omega-\omega_Q)^2+\Gamma^2} + \frac{\Gamma}{(\omega+\omega_Q)^2+\Gamma^2}\right] \quad , \tag{2}$$

and the damped harmonic oscillator (DHO),

$$F_Q^t(\omega) = \frac{\Gamma}{\pi}\frac{\omega_Q^2}{(\omega^2-\omega_Q^2)^2+\omega^2\Gamma^2} \quad . \tag{3}$$

To analyze the quasielastic peak which appears below $T = 100$ K we used a Lorentzian shape, namely,

$$S_e(Q,\omega) = \frac{I}{\pi}\frac{\Gamma_0}{\omega^2+\Gamma_0^2} \quad , \tag{4}$$

yielding two supplementary parameters I and Γ_0 which describe, respectively, the integrated intensity and the linewidth. The parameters were then adjusted in order to get the best fit to the data. The lines drawn in Fig. 1 are the results of the convolution of Eq. (1) with the spectrometer resolution using the DHO form to $F_Q^t(\omega)$. (The choice of the DL form gives fits of similar quality.) As for the spin waves a comparison of the results obtained for different Q values at a given temperature shows that a quadratic law $\omega_Q = DQ^2$ accounts for the data in the whole temperature range.[18]

The temperature dependence of the stiffness constant evaluated through the DHO and the DL models is shown in Fig. 2(a). The two fits yield different values of D when the spin-wave linewidths are comparable to their energy [see Fig. 2(b)]. When using a DL model we observe a decrease of D between 180 and 80 K which corresponds to the observations in other systems[11,12] using the same model. However, in the same temperature range the choice of the DHO form yields a constant D value as already noticed.[19] In the present case, as distinct from all previous studies,[11,12,19] further lowering of temperature below ~70 K shows a remarkable increase of D. It is important to note that this increase in D is seen irrespective of the spectral form used in the analysis. The spin-wave linewidth Γ [Fig. 2(b)] reaches a constant minimum value between 180 and 240 K. At higher temperatures Γ increases as expected in a FM when approaching T_c but Γ also increases progressively at lower temperatures and reaches a maximum value somewhere between 60 and 100 K. At a given temperature, Γ increases rapidly with Q. However, because of the limited Q range of the data, no simple law relating Γ and Q could be established with certainty. An anomalous increase of the linewidth has also been observed in the Fe-Cr and amorphous Fe-Ni systems.[11,12] The static transverse susceptibility χ_Q^t obeys a $1/Q^2$ law at all temperatures in the experimental-Q range, as in usual FM, but shows an anomalous temperature dependence [Fig. 2(c)]. The quantity $N = \chi_Q^t \cdot Q^2$ which is usually constant below T_c, progressively increases in the present case below 180 K and reaches a constant value below 100 K.

Another interesting feature of the data is the appearance of quasielastic scattering below ~100 K which we denote as T_f' (to be compared with $T_f \sim 40$ K, the reentrance temperature). The linewidth of the quasielastic scattering decreases as T is lowered and becomes resolution limited below 40 K. Its integrated intensity increases when the

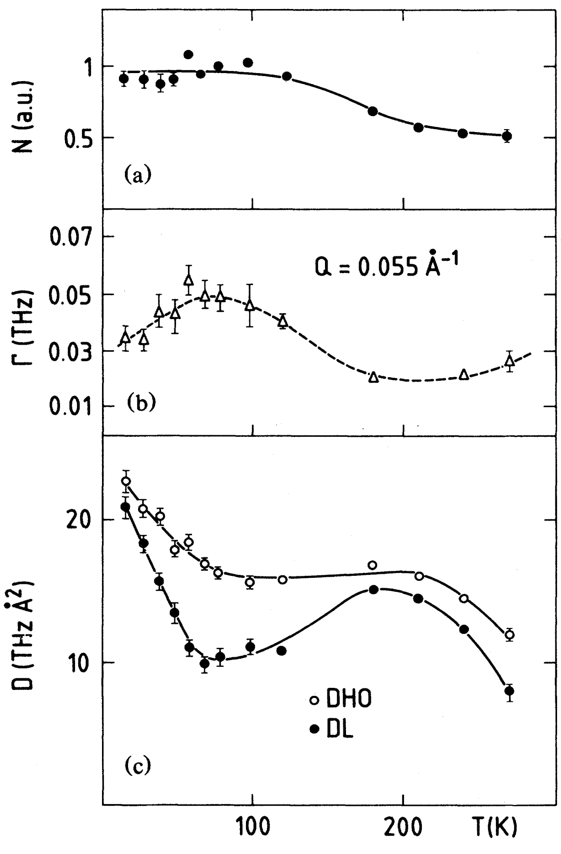

FIG. 2. Temperature dependence of (a) the transverse static susceptibility as deduced from the fits using the DHO model and assuming $\chi_Q^t = N/Q^2$, (b) the spin-wave damping for $Q = 0.055$ Å^{-1} (DHO model), and (c) the stiffness constant evaluated through the damped harmonic oscillator (DHO) and the double Lorentzian (DL) model. The lines are guides to the eyes. The error bars, when not indicated, are included in the size of data points.

temperature is lowered. As already pointed out for the other reentrant systems,[7,8] the simple law $I \propto (Q^2+K^2)^{-1}$, where K is the inverse of a correlation length, cannot account for the Q dependence of the integrated intensity. In the present case, the experimental Q range is too restricted to ascertain any other law.

The present results call for some comments on similar experiments in Fe-Cr (Ref. 11) and amorphous Fe-Ni (Ref. 12), where no spin waves were detected at low temperatures. A significant difference is perhaps that the stiffness constant D in these systems is about half that in the present $Ni_{78.4}Mn_{21.6}$ alloy. The nonobservation of spin waves below T_f is possibly related to the fact that measurements were made at Q values for which the spin waves are strongly damped. For example, in the present experiment, no resolved spin waves could be detected below 100 K for $Q \leq 0.075$ Å^{-1}.

In conclusion, we have observed well-defined spin waves in a reentrant ferromagnet at all temperatures below T_c (330 K) particularly and, for the first time, below T_f, the temperature of the down turn in the ac susceptibility which is thought to mark the reentrant temperature. The spin waves coexist with a quasielastic scattering which appears below a temperature T_f' (~ 100 K) which is higher than T_f (~ 40 K). At some intermediate temperature (~ 70 K) between T_f' and T_f (neither of which are sharp) the spin-wave stiffness constant shows a marked increase with decreasing temperature, a feature which may or may not be particular to Ni-Mn alloys. Thus, although the spin dynamics reveals some anomalous features, namely, an increase of Γ and $\chi^t(Q)$ below 180 K and an increase of D below 70 K, it clearly contradicts the common assumption of a breakdown of ferromagnetism at T_f made in earlier studies.[11,12] The persistence of ferromagnetic correlations is corroborated by resistivity measurements which probe spin correlations on the scale of the electron mean free path and show no anomaly either at T_f' or at T_f.[20] It is plausible that the so-called reentrant state is simply a coexistence of SG and FM ordering as suggested by Gabay and Toulouse.[4]

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