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Magnetic structure observed in an applied field in reentrant $Au_{0.81}Fe_{0.19}$ and $Ni_{0.81}Mn_{0.19}$ single crystals

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Spin correlations have been studied in two reentrant single crystals, $Au_{0.81}Fe_{0.19}$ and $Ni_{0.81}Mn_{0.19}$, by small-angle neutron scattering at low temperature (11 K) and in applied fields in the range of 1.5 to 8 kOe. They reveal the existence of a magnetic structure, in all **q** directions, which can be attributed mainly to transverse spin components. These results, which are insensitive to the crystal orientation, corroborate previous observations in polycrystalline $Ni_{0.78}Mn_{0.22}$ and amorphous $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ systems. The overall similarities found in these three systems, which have very different nuclear structure or a different kind of chemical short-range order, are very remarkable. Measurements of the elastic scattering contribution carried out in the 11 to 200 K temperature range at constant field on $Ni_{0.81}Mn_{0.19}$ reveal that the structure disappears well below T_c at a temperature which could be assigned to the canting transition of mean-field theory.

The observation of an isotropic magnetic structure in the presence of an applied field in "reentrant" metallic polycrystalline $Ni_{0.78}Mn_{0.22}$ and amorphous $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ (Refs. 1 and 2) has raised several questions which have led us to perform further small-angle neutron scattering experiments. The choice of single crystals $Ni_{0.81}Mn_{0.19}$ and $Au_{0.81}Fe_{0.19}$ is aimed at checking whether the polycrystalline state introduces some spurious effects related to grain average. The study of a third system, $Au_{0.19}Fe_{0.81}$, very well characterized by other techniques, was undertaken in order to confirm the generality of this magnetic structure. Moreover, comparison between $Ni_{0.19}Mn_{0.81}$ and $Au_{0.19}Fe_{0.81}$ was of special interest since they are known to exhibit chemical short-range orders (SRO) of very different kind (tendency to Ni_3Mn order in the first case,[3] and to the formation of Fe-rich platelets in the second[4] case). Hence the influence, if any, of the SRO may be determined. The present experiments reveal the existence of a similar structure in $Au_{0.81}Fe_{0.19}$ which supports the generality of these observations in the reentrant spin-glass state. The temperature dependence of magnetic correlations at constant field has only been studied in $Ni_{0.81}Mn_{0.19}$, since the strong neutron absorption of Au would require prohibitively long counting times for $Au_{0.81}Fe_{0.19}$.

We report here neutron scattering measurements obtained by two distinct techniques: diffraction experiments at small angle (SANS) and elastic experiments. The SANS diffraction experiments are only meaningful when the contribution of the inelastic scattering to the measured intensity is negligible, i.e., only at the lowest temperature (11 K) in the present case. The measurements have been carried out on the D11 spectrometer at the Institut Laue Langevin ILL reactor using a XY multidetector. Scattered wave vectors in the range $10^{-2} < q < 1.3\ 10^{-1}$ Å^{-1} were obtained with an incident wavelength of 5.5 Å and sample to detector distances of 5 and 2.5 m. The $Ni_{0.81}Mn_{0.19}$ and $Au_{0.81}Fe_{0.19}$ samples were, respectively, 10 and 0.8 mm thick and had no specific orientations with respect to the field, but orientation effects were checked as explained below. The data have been corrected in the usual way for sample environment scattering and detector efficiency, and gathered in sectors of 30° in the detector plane. The intensities have been put in b sr^{-1} atom^{-1} by reference to the incoherent nuclear scattering of a slab of nickel single crystal. The elastic experiments enable separation between the elastic component and the spin waves, the contribution of which increases with temperature. Elastic measurements have been carried out on the three-axis spectrometer 4F2, at the Orphée Reactor (LLB) with 2.28 meV of incident energy and 60′-25′-20′-20′-20′ of collimation resulting in an energy resolution of 15 μeV. The magnetic field was applied parallel to the scattering vector direction. Data were background corrected by subtracting temperature-independent spectra obtained between 120 and 200 K, i.e., in the ferromagnetic state (cf. below). The present measurements were carried out between 11 and 200 K in applied fields up to 8 kOe. The characteristic temperatures were determined from susceptibility measurements in our samples. The Curie temperatures are 260 and 160 K for $Ni_{0.81}Mn_{0.19}$ and $Au_{0.81}Fe_{0.19}$, respectively. The spin-glass freezing temperatures T_f can be deduced from the inflection point in the low-temperature decrease of the susceptibility which corresponds to a maximum of the imaginary susceptibility $\chi''(T)$ (Ref. 5) and evaluated to about 30 and 25 K in $Ni_{0.81}Mn_{0.19}$ and $Au_{0.81}Fe_{0.19}$, respectively. As already mentioned, the dominant feature of the observations is the

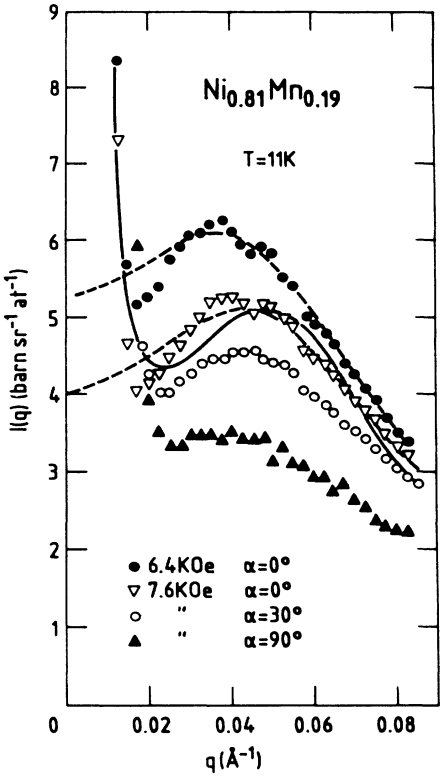

FIG. 1. $Ni_{0.81}Mn_{0.19}$: $I(q)$ intensities at 11 K for $H=6.4$ kOe, $\alpha=0$ (●) and $H=7.6$ kOe, $\alpha=0$, 30, 90 (▽,○,▲). The continuous line is the best fit obtained using the sum of a double Lorentzian (DL) function and a $Aq^{-\alpha}$ contribution. The dashed lines are best fits obtained with DL only using data truncated at $q=0.025$ Å^{-1} (see text).

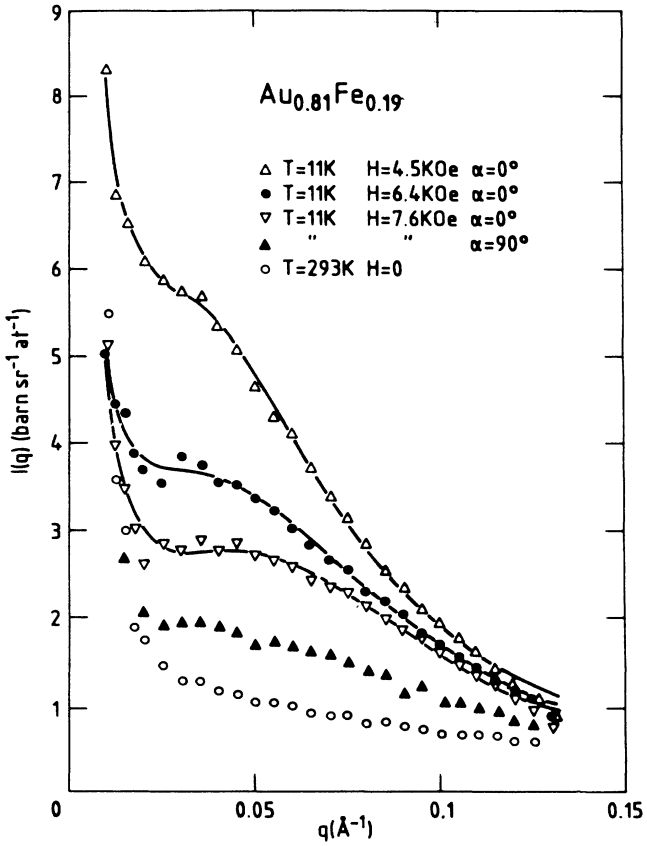

FIG. 2. $Au_{0.81}Fe_{0.19}$: $I(q)$ intensities at 11 K for $\alpha=0$ at $H=4.5$, 6.4, and 7.6 kOe (△,●,▽) and for $\alpha=90$ at 7.6 kOe (▲). The continuous lines are best fits obtained with a double Lorentzian function and a $Aq^{-\alpha}$ contribution. $I(q)$ at $T=290$ K ($2T_c$) is plotted for comparison (○).

existence of a maximum in $I(q)$, the position of which is isotropic but strongly field dependent, while the intensity is a function of $\alpha=(\mathbf{q},\mathbf{H})$, the angle between the scattered wave vector and the field. We examine below first the q and α dependence of the scattering at 11 K, and then its temperature dependence (for the *Ni*Mn case only).

(1) *q-dependence analysis of the scattering.* Figures 1 and 2 illustrate the observed scattering $I(q,\alpha=0)$ as a function of the field at $T=11$ K for both systems. A well-defined maximum is observed in *Ni*Mn while in *Au*Fe the intensity looks rather like a shoulder on a strong q-dependent contribution at the smallest q values ($q \leq 0.02$ Å^{-1}). The presence of similar features at any angle is also illustrated in Fig. 1 ($\alpha=30°$ and 90°) and Fig. 2 ($\alpha=90°$), even if slightly smeared out by the decrease of the intensity when α increases up to 90°. A precise analysis of this structure, especially in $Au_{0.81}Fe_{0.19}$, is impeded by the existence of the strong low-q scattering. The latter scattering may have different origins nuclear as well as magnetic (surface defects, dislocations, etc.). Its importance with respect to the magnetic signal of interest is most likely due to the small volume/surface ratio of the $Au_{0.81}Fe_{0.19}$ sample, necessitated by the strong neutron absorption of Au. A measurement at $T=2T_c$ reveals the persistence of magnetic scattering, thus preventing direct measurement of the temperature-independent low-q nuclear contribution. The incoherent scattering (isotopic, chemical, magnetic) has been evaluated in both systems to less than one hundred of millibarns, and is therefore negligible. Furthermore, in both cases a rotation of the single crystals around the incident neutron axis revealed that the low-q intensity is slightly dependent on the crystallographic direction. On the contrary, the magnetic structure appearing at larger q is independent of the sample orientation and for $q \geq 0.02$ Å^{-1}, the observations in $Ni_{1-x}Mn_x$ are completely similar in the single crystal and the polycrystalline samples. A subtraction of high-temperature scattering as done previously[1,2] being unsatisfactory, because of the residual magnetic scattering, we tried to account for it in the data analysis by an empirical modelization in an $Aq^{-\alpha}$ law. The larger q contribution was tentatively described by a double Lorentzian (DL),

$$B\left[\frac{\Gamma}{\Gamma^2+(q-q_0)^2}+\frac{\Gamma}{\Gamma^2+(q+q_0)^2}\right],$$

as in Ref. 6. A constant term added to account for a possible incoherent scattering was found to be negligible in all cases. In the $Ni_{0.81}Mn_{0.19}$ system, where the structure

is well shaped, this parametrization was unable to describe the data correctly. An alternative analysis consists in considering only data for $q \geq 0.025$ Å^{-1}, modeling them by a DL only. It yields fits of better quality but with a systematic discrepancy as the low-q intensity is found weaker than expected from Lorentzian law (see Fig. 1). (This discrepancy would be still larger if there remained a slight contamination of the neglected low-q part.) The same observations were made in polycrystalline $Ni_{0.78}Mn_{0.22}$ and amorphous Fe-Mn-P-B-Al, indicating a systematic asymmetry in the structure shape. We reach the same conclusions using a double-squared Lorentzian law instead of DL functions. We notice that the squared Lorentzian law yields much better fit at large q ($q > 0.1$ Å^{-1}) than the Lorentzian law. In the case of $Au_{0.81}Fe_{0.19}$, we obtain good fits with the sum of a double Lorentzian and a $Aq^{-\alpha}$ law (Fig. 2). However, this system, for which the magnetic structure is more hidden by the low-q part, as mentioned above, cannot provide a good characterization of the real structural shape. The bad fits at low q prevent us from drawing any physical meaning to Γ values. However, the deduced q_0 and B values corroborate the previous qualitative observations: namely, the decrease of the intensity at the maximum and the shift of the position of the maximum toward larger q values as the field increases. It should be noticed that in the range 2 to 8 kOe, where the experiments have been performed, the magnetization varies only a few percent: Thus the observed decrease of the magnetic intensity with field is induced by a change in the correlations, and cannot be explained by a change in the mean spin components.

(2) *α dependence of the scattering.* In all the systems studied the magnetization measurements indicate an apparent saturation of the magnetization at rather low fields (about 1.5 to 2 kOe), allowing a meaningful distinction between transverse and longitudinal correlations to be made. The magnetic scattered intensity may be written from the general neutron cross-section

$$I(q) \propto \int_r \int_{r'} \langle M_r^{\perp} M_{r'}^{\perp} \rangle e^{iq(r-r')} d^3r\, d^3r' ,$$

with the single assumption of an uniaxial symmetry:

$$I(q,\alpha) = \cos^2\alpha 2T(q,\alpha) + \sin^2\alpha [T(q,\alpha) + L(q,\alpha)] , \quad (1)$$

where $T(q,\alpha)$ and $L(q,\alpha)$ are the Fourier transforms of the correlation functions of spin components perpendicular and parallel to the mean magnetization, respectively. In Fig. 3 the α dependence of the intensity at some q values and H values for both samples is reported. Except in the low-q range ($q \leq 0.03$ Å^{-1}) the intensity follows fairly well an $A\sin^2\alpha + B\cos^2\alpha$ law, even though a slight systematic discrepancy is observed in all samples. This implies that the spin correlations (T and L) are nearly α independent: i.e., isotropic in real space. If we look at the ratio

$$R(q) = \frac{I(q,\alpha=0)}{I(q,\alpha=90°)} = \frac{2T(q)}{L(q)+T(q)} ,$$

we find that beyond a minimum field value which corresponds to the "apparent saturation" field in the magnetization curve, this ratio does not vary with the field, keeping the value of about 1.55 (to be compared to the value of

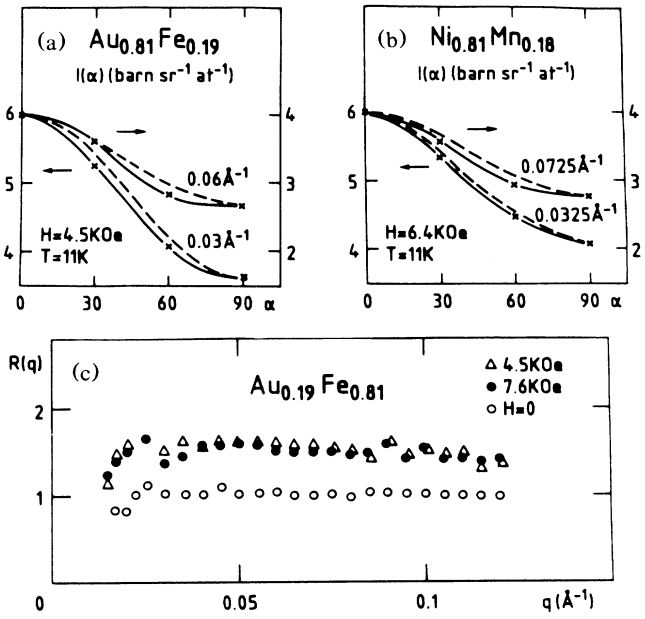

FIG. 3. $I(\alpha)$ at constant q, for typical q and H values. The continuous lines are a guide for the eye. The dashed lines are a fit using a $A\sin^2\alpha + B\cos^2\alpha$ law [Eq. (1) in the text]. (a) $Au_{0.81}Fe_{0.19}$, (b) $Ni_{0.81}Mn_{0.19}$, (c) anisotropy ratio $R(q)$ at zero field and two field values in $Au_{0.81}Fe_{0.19}$.

1.7 found when high-temperature data are subtracted as was done previously[2]). This means that the longitudinal part is less than $\frac{1}{4}$ of the total scattering ($\alpha = 90°$) and indicates that the observed structure is dominated by a specific arrangement of the transverse components rather than by a modulation of the longitudinal ones, which would yield $R < 1$. But the longitudinal correlations clearly contribute to the observed structure as they vary with the field in the same way as the transverse correlations (R is H independent). We emphasize that the R value is not directly related to the ratio between the mean longitudinal and transverse spin lengths, i.e., the canting angle. This last quantity can be deduced, for instance, from the comparison between the "saturation" magnetization value, or the magnetic Bragg intensities (S_z^2) and the Mössbauer data[7] (related to S^2).

(3) *Temperature dependence.* The temperature dependence of the magnetic correlations has been measured between 11 and 200 K at 5.8 and 7.8 kOe on the $Ni_{0.81}Mn_{0.19}$ single crystal. The scattered intensity at constant field strongly decreases as temperature increases up to about 120 K. From 120 K up to the highest temperature studied (200 K), the small measured signal is temperature and field independent and no more maximum can be seen in the q dependence. It is therefore identified as the sample background and subtracted from the raw data. Of course this procedure cannot be used in the diffraction technique because of the spin-wave contribution. Corrected data at several temperatures are shown in Fig. 4. They show the same features as those observed from the diffraction experiments at 11 K, namely, a maximum in the $I(q)$ scattering at some q_{max} value which moves to-

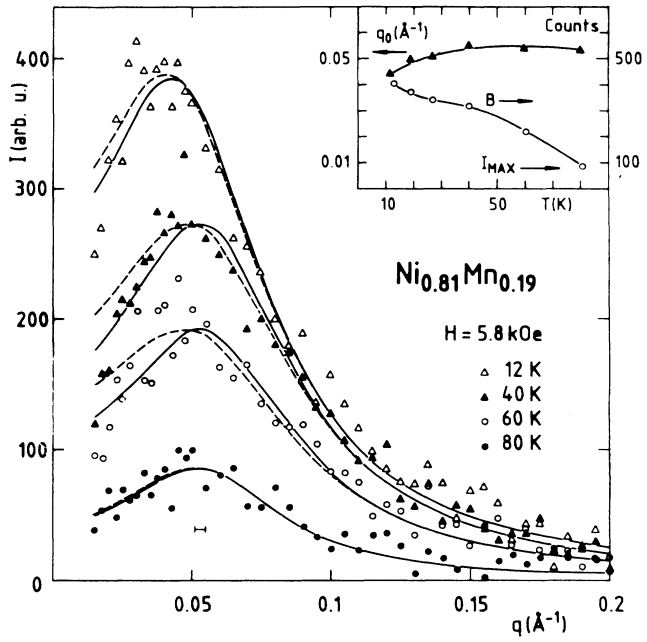

FIG. 4. $Ni_{0.81}Mn_{0.19}$: Elastic scattering intensities $I(q)$ measured at 5.8 kOe for various temperatures. Continuous lines are DL fits with considering all data and dashed lines with data truncated at $q=0.03$ Å^{-1}. The bar represents the q resolution. In inset are reported the temperature dependence of q_0 (▲) and the q integrated intensity B (○) deduced from fits with considering all data. The continuous lines of the inset are guides for the eye.

ward higher value when the field increases. The q value of the intensity maximum slightly decreases as the temperature increases at constant field. This appears for instance in the Lorentzian parametrization of the curves (see the inset).

In conclusion, the overall similarity of the magnetic correlations found in three systems, polycrystalline and single-crystal $Ni_{1-x}Mn_x$, single-crystal $Au_{0.81}Fe_{0.19}$, and amorphous (Fe-Mn-P-B-Al) of very different nuclear structure and chemical short-range order is very remarkable. Other recent data[6,8] confirm some universal behavior of "reentrant" systems. The transverse character of this magnetic structure can be ascertained at low temperature. In $Ni_{0.81}Mn_{0.19}$ the temperature of 120 K which marks the occurrence of this transverse structure in applied fields is also the temperature which locates the increase of a strong elastic scattering in zero field. See, for instance, elastic measurements in the sample $Ni_{0.784}Mn_{0.216}$ (Ref. 9) which have very similar transition temperature to the present one. In Fe-Mn-P-B-Al (Ref. 2) the temperature at which the small-q scattering occurs has been identified with that for the anomalous increase of the mean hyperfine field determined from Mössbauer spectra at $H=0$ and assigned to the canted state of mean-field theory.[7] The present observations strengthen this interpretation. We can readily think that the decrease of the intensity with temperature at constant field observed for this magnetic scattering corresponds to a decrease of the transverse spin lengths in relation to that of the q_T order parameter of mean-field theory.[10] The "canting" temperature T_K for transverse spin-component freezing (for instance 120 K in $Ni_{0.81}Mn_{0.19}$) is much higher than the longitudinal freezing temperature T_f determined for instance from susceptibility measurements ($T_f=30$ K in $Ni_{0.81}Mn_{0.19}$ as mentioned above). We believe that these two temperatures are likely related to two distinct phenomena (as predicted by Ref. 10) rather than a single one observed with different time windows. The transverse components were already known to be correlated in zero field as proved by the q-dependent scattering at $H=0$ and by recent ^{197}Au Mössbauer experiments.[11] We show in this paper that they exhibit a very specific arrangement in applied fields yielding a maximum in $I(q)$. The origin of this magnetic structure remains to be understood.

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