Diffuse satellite peaks in a ferromagnetic $Pd_{97.5}Mn_{2.5}$ single crystal

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A ferromagnetic $Pd_{97.5}Mn_{2.5}$ alloy single crystal was studied by neutron scattering and susceptibility measurements. The diffuse satellite peaks along the [1 0 0] axis were observed for the ferromagnetic alloy. The diffuse satellite peaks, however, disappeared under a magnetic field of 5 T applied along the [0 0 1] direction perpendicular to the (0 0 1) scattering plane. Combined with the susceptibility data, we discuss several models for the ferromagnetic state of the dilute PdMn alloy.

I. INTRODUCTION

The magnetic phase diagram of fcc PdMn alloy is rather mysterious. The alloy with a Mn concentration below 4 at. % shows ferromagnetism although the maximum Curie temperature ($T_c = 7$ K) appears at 2.5 at. % Mn. The alloy with a Mn concentration higher than 5 at. % behaves like a spin glass.[1] Many experimental data have been reported for ferromagnetic PdMn alloys and the similarity with the enhanced ferromagnetic systems PdFe and PdCo has been pointed out.[2–4] In contrast to the PdFe alloy, however, as the Mn concentration increases, the direct *d-d* interaction which couples antiferromagnetically with the nearest-neighbor Mn-Mn pair becomes predominant for the PdMn alloy. To explain this mysterious magnetic phase diagram, previous authors proposed the following model.[5] The nearest-neighbor Mn-Mn pairs couple antiparallel, but the second-neighbor pairs favor ferromagnetic coupling through polarization of the conduction electrons. For the low Mn concentration alloys, the probability that two Mn atoms occupy nearest-neighbor sites is very low and the ferromagnetic coupling is predominant. As the Mn concentration increases, the nearest-neighbor probability increases and the conflict between first- and second-neighbor interactions prevails. This causes the system to exhibit a spin-glass phase. Thus, the PdMn alloy has been regarded as a non-Ruderman-Kittel-Kasuya-Yosida (RKKY) spin glass.

Very recently, however, one of the present authors (Y.T.) and co-workers[6] observed the diffuse satellite peaks in neutron-scattering experiments on PdMn spin-glass alloys with Mn concentrations of 10 and 20 at. % and revealed that the spin-glass-like behavior comes from the process of freezing in dynamically fluctuating spin-density wave (SDW) clusters as is the case for CuMn spin-glass alloys.[7] The SDW propagates along the [1 0 0] axis and the wavelength of the SDW varies with the Mn concentration. Various experimental data support the idea that the SDW in PdMn alloys is a reflection of the special shape of the Fermi surfaces of the alloy, suggesting that the RKKY interaction plays an essential role in its spin-glass-like behavior. Furthermore, the wavelength of the SDW extrapolated to the limit of low Mn concentration is approximately twice the lattice parameter [$Q_{SDW} \sim (0.5,0,0)$]. This indicates that the second-neighbor Mn-Mn pair favors antiferromagnetic coupling. This is inconsistent with the model proposed by the previous authors.

The purpose of the present work is to solve the mystery of the magnetic phase diagram of PdMn alloys using neutron scattering and susceptibility measurements. To execute this aim, a 2.5 at. % Mn alloy was chosen for two reasons: (1) The maximum Curie temperature in the ferromagnetic phase occurs at this Mn concentration; and (2) The extrapolation of the concentration dependence of the spin-glass freezing temperature to 0 K crosses around 2.5 at. % Mn.[1] For this sample, the susceptibility data actually showed a ferromagnetic response, but the diffuse satellite peaks were observed in the neutron-scattering experiment on this same specimen. Further neutron-scattering measurements with an applied magnetic field were performed to investigate the interrelation between the ferromagnetism and antiferromagnetic satellite reflections. We propose several possible models to explain the apparently incompatible features of the data obtained by these two different experimental methods.

II. SAMPLE PREPARATION AND EXPERIMENTS

A single crystal of $Pd_{97.5}Mn_{2.5}$ with a volume of about 2 cc was grown in a furnace with a carbon heater system under an Ar atmosphere. A small part of the single crystal was cut out from the ingot for the susceptibility measurements and the rest was used for the neutron-scattering experiments. The specimen was furnace cooled with a cooling rate of about 300 °C/min. around 1000 °C.

Susceptibility data were taken using a superconducting quantum interference device system at the Materials Characterization Central Laboratory, Waseda University. Preliminary neutron-scattering measurements were performed at the $T1$-1 triple axis spectrometer installed on a thermal guide of JRR-3M, Tokai and the final data were taken on the HB-3 triple axis spectrometer installed at the HFIR, Oak Ridge. All of the data were taken in the elastic-scattering mode using $E_0 = 13.5$ meV and 14.75 meV incident neutrons for $T1$-1 and HB-3, respectively, and a pyrolitic graphite analyzer. The energy resolution of the instrument configuration used is estimated to be $\sim$0.65 meV full width at half maximum, indicating that the spin motion with a characteristic time shorter than 10^{-11} s is discarded by rejecting inelastic-scattering processes.

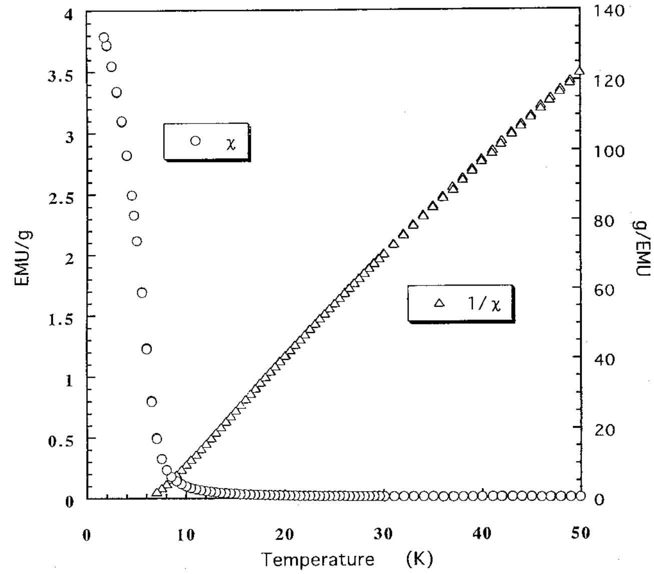

FIG. 1. Temperature variation of the magnetization studied under a field of 200 Oe together with the inverse susceptibility above T_c.

III. EXPERIMENTAL DATA

The temperature variation of the magnetization studied at 200 Oe is given in Fig. 1. The magnetization data are typical of ferromagnetic materials and the Curie temperature is estimated to be 7 K. This value is consistent with the phase diagram reported by Ho *et al.*[1] The field dependence of the magnetization has also been studied up to 5 T at 3 and 10 K and the results are given in Fig. 2. The maximum field is not sufficient to achieve saturation magnetization, which is consistent with previous reports. Star *et al.* reported that a magnetic field of 21 T is still not enough to saturate the magnetization of a $Pd_{97.55}Mn_{2.45}$ alloy.[8] The magnetization curve suggests that the magnetization process proceeds by two

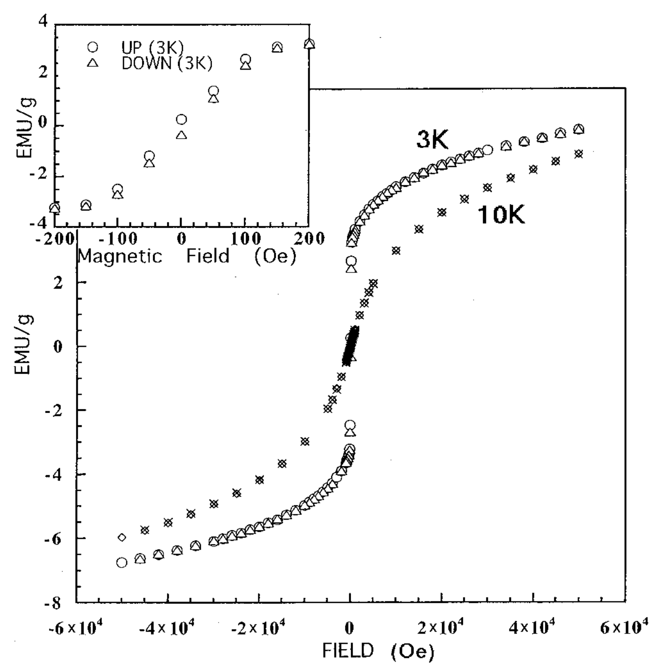

FIG. 2. Magnetization curve studied at 3 and 10 K. The different symbols show data for increasing and decreasing field. The inset is an enlarged view of the hysteresis loop at 3 K.

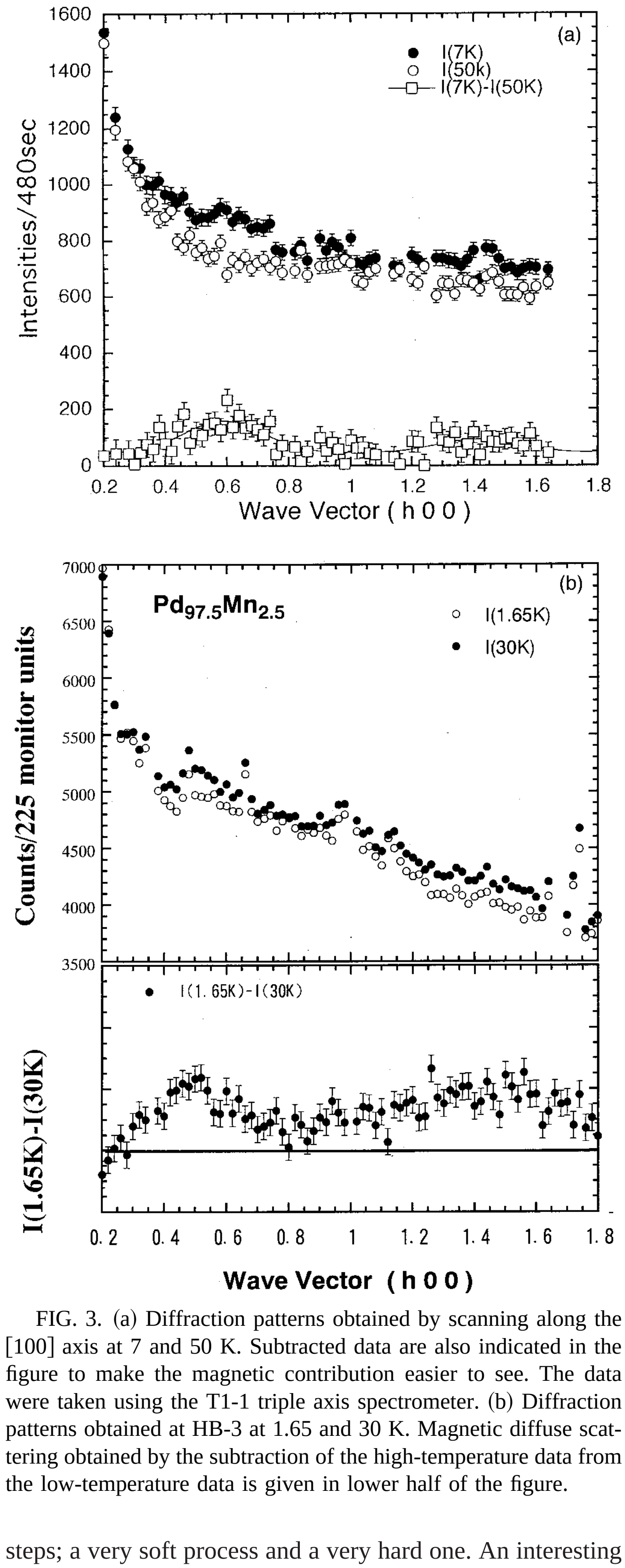

FIG. 3. (a) Diffraction patterns obtained by scanning along the [100] axis at 7 and 50 K. Subtracted data are also indicated in the figure to make the magnetic contribution easier to see. The data were taken using the T1-1 triple axis spectrometer. (b) Diffraction patterns obtained at HB-3 at 1.65 and 30 K. Magnetic diffuse scattering obtained by the subtraction of the high-temperature data from the low-temperature data is given in lower half of the figure.

steps; a very soft process and a very hard one. An interesting feature is observed for the hysteresis loop as shown in the inset of Fig. 2. The hysteresis loop is very narrow and closes at a low field of about 200 Oe. One of the noticeable features is that the magnetization process is reversible. This feature is contrasted with the spin-glass alloys for which strong memory effects are observed below the freezing temperature.

Neutron-scattering line profiles obtained by scanning

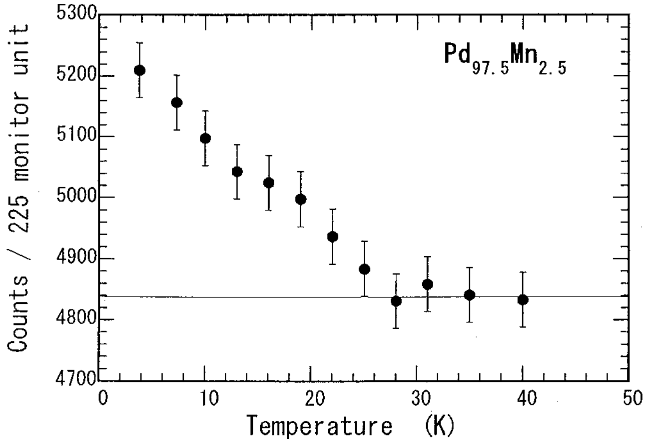

FIG. 4. Temperature variation of the diffuse satellite peak intensity.

along the [1 0 0] direction passing through the 1 0 0 reciprocal-lattice point are shown in Fig. 3. Figure 3(a) shows the data taken at the $T1$-1 spectrometer at 7 and 50 K, and their difference, $I(7\,\mathrm{K})$-$I(50\,\mathrm{K})$. In Fig. 3(b), the subtracted data taken at the HB-3 at HFIR using the same specimen are given. These subtracted data show diffuse satellite peaks at around $1\pm\delta\,0\,0(\delta\sim0.5)$, indicating that the SDW clusters still exist in this ferromagnetic alloy. These diffuse satellite peaks are very similar to those observed in concentrated Mn alloys which show typical spin-glass-like susceptibility, although the satellite peaks of the present data are ill-defined due to low scattering intensity. The wave vector of the SDW is estimated to be 0.5 in reciprocal-lattice unit $(2\pi/a)$. Thus the RKKY interaction plays an essential role even at this concentration and the data suggest that the second-neighbor Mn atoms couple antiferromagnetically.

The temperature variation of the diffuse satellite peak intensity studied at $Q=0.56$ (rlu) is given in Fig. 4. The satellite peaks disappear around $T\sim27$ K (±2 K) and no anomaly in the peak intensity is observed at $T_c=7$ K.

In order to examine the relation between the ferromagnetic susceptibility and the diffuse satellite peaks observed by neutron diffraction, the magnetic diffuse satellite peaks were studied under an applied magnetic field at 1.7 K. A magnetic field of 5 T was applied along the direction perpendicular to the (0 0 1) scattering plane. To render the magnetic-field effect more conspicuous, the difference between the data with the applied magnetic field and those without the magnetic field is plotted in Fig. 5. Careful study of the structure of the difference data indicates that the diffraction pattern in Fig. 5 is very similar to that in Fig. 3(b) and that the background counts increased when the magnetic field was applied. In order to aid the reader in interpretating the diffraction profiles, the various components of the scattering are schematically illustrated in Fig. 6. Thus, by applying a magnetic field of 5 T, the diffuse satellite peaks faded away and the background intensity increased. To confirm the effect of the applied field, the difference between the data at 30 K where the diffuse satellite peaks disappear and those at 1.7 K under the magnetic field is plotted in Fig. 7. The figure shows a plateaulike feature and supports disappearance of the diffuse satellite peaks when the 5 T magnetic field is applied.

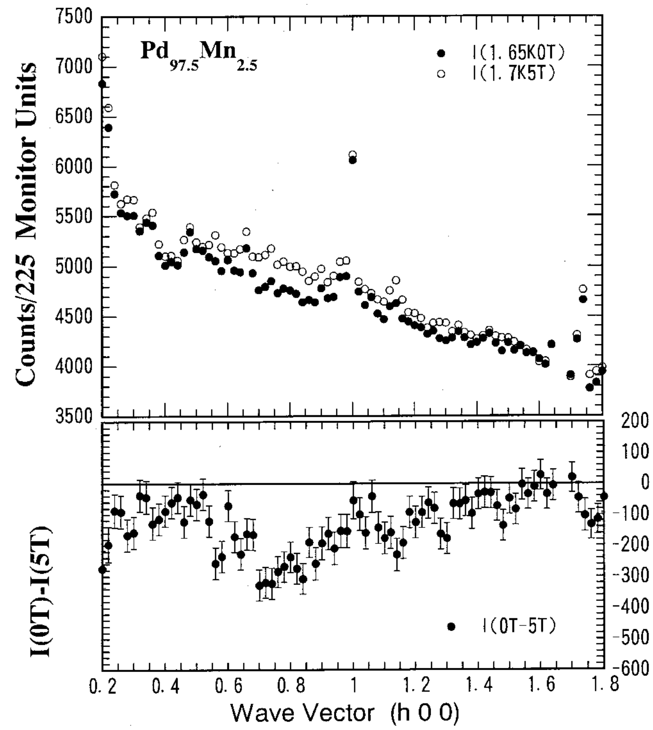

FIG. 5. Diffraction patterns obtained with and without magnetic field. Difference plot of the data under a magnetic field of 5 T and those taken without the magnetic field is given in lower half of the figure.

IV. DISCUSSION

Although the Curie temperature, determined by the susceptibility measurements is 7 K, the diffuse satellite peaks disappear at around 27 K. This does not necessarily mean that the ferromagnetic region and the SDW regions are spatially separated because susceptibility and neutron-scattering measurements observe responses with different time scales. In metallic spin glasses, this phenomenon is very common. The diffuse magnetic peaks observed by neutron scattering usually survive up to far above the freezing temperature at which the susceptibility data show a cusp-type anomaly. This is due to the difference in the characteristic times of these experimental methods with a rather wide time resolution window for neutron-scattering experiments.

The susceptibility data show that the magnetization curve undergoes two step changes, a very soft component which saturates about 200 Oe and a very hard component which does not saturate even in the 5 T field. Note that the hard component shows behavior similar to the magnetization curve observed above T_c.

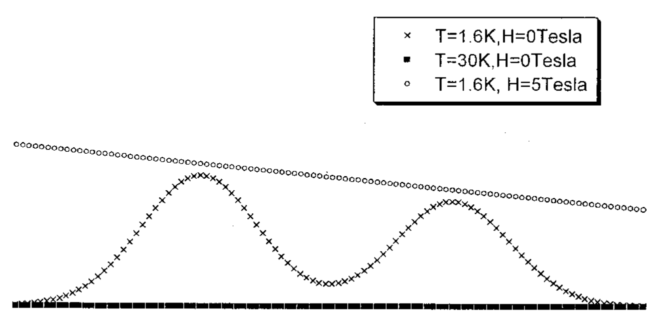

FIG. 6. Schematic illustration of the scattering intensities studied under the various conditions.

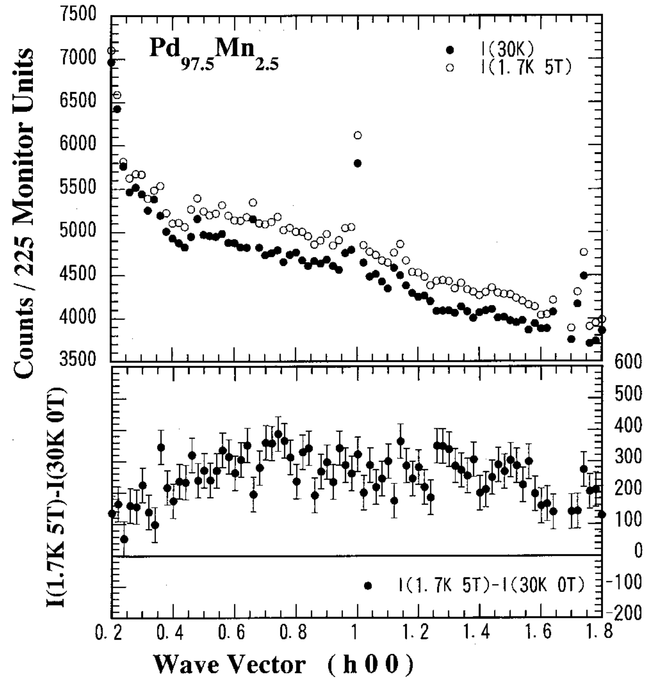

FIG. 7. Observed line profiles studied at 30 K without a magnetic field and at 1.7 K with an applied magnetic field of 5 T. Difference of these data is also given.

In order to explain the incompatible features between the ferromagnetic susceptibility and the diffuse satellite peaks in neutron scattering, three possible models are considered.

(1) Inhomogeneous clusters model: In this model, a specimen is composed of small clusters with different magnetic phases. Some clusters would show ferromagnetism and others do the spin-glass phase (SDW clusters). The volume fraction of the SDW clusters can be estimated using the satellite peak intensities for the 2.5% Mn alloy and those for the 15% Mn alloy under the assumption that the satellite intensity is proportional to the number of Mn-Mn pairs and the magnitude of the Mn moment does not depend on the Mn concentration. The estimated volume fraction of the SDW clusters is about 80% of the total volume. On the other hand, the ferromagnetic volume fraction is estimated to be several % using the observed saturation magnetization of the soft component and the moment value determined from the observed Curie constant. These values are not surprising. Previous authors suggested that an antiferromagnetic spin correlation plays a rather important role for ferromagnetic PdMn alloys with relevant Mn concentrations.[4,8] Star *et al.* estimated from their susceptibility and specific-heat data that the fraction of Mn atoms with antiferromagnetic coupling is about 60% for a 2.45 at. % Mn alloy.[8]

The susceptibility data obtained here are completely explained by this inhomogeneous cluster model. Since the ferromagnetic cluster sizes are rather small, they would exhibit superparamagnetism with a very low Curie temperature. The free ferromagnetic clusters would be first to respond to a very small magnetic field resulting in the soft magnetization process. The observed Curie temperature (7 K) would be this process. The hard magnetic behavior remains even above the Curie temperature because other ferromagnetic clusters, which couple through the SDW clusters, respond to a higher magnetic field. Within this model, the effect of the magnetic

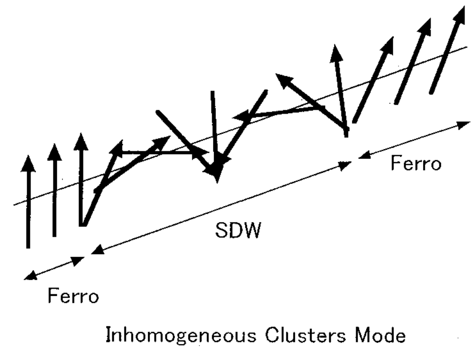

FIG. 8. Inhomogeneous cluster model in which ferromagnetic clusters couple through the SDW clusters.

field on the neutron-scattering data observed here can be explained as follows. We assume that most of the ferromagnetic clusters couple through the short-range SDW clusters as shown in Fig. 8. With no applied field ($H=0$ T), the ferromagnetic moments of the clusters would point in various directions. When a magnetic field is applied, the moments of the ferromagnetic clusters tend to orient parallel to the applied field. Thus the moments of the SDW clusters become twisted and the wave vector of the SDW becomes ill-defined. Since the twisting angle of each SDW cluster would be different, the period of the SDW in the clusters would be broadly distributed. Thus, the satellite peaks disappear and the background counts along the [1 0 0] direction increase. However, this model does not explain how the ferromagnetic clusters are stabilized in the low Mn concentration alloy. The Mn concentration of the present specimen is considered to be rather homogeneous, otherwise the Curie temperature should be lower than 7 K from the reported magnetic phase diagram.[1]

(2) Transversely modulated ferromagnetic model: Many previous authors pointed out the similarity between the ferromagnetism of PdMn alloys and the enhanced ferromagnetism found in PdFe alloys. Recent neutron-scattering measurements on PdFe, PtFe, PdCo, and PtCo alloys, all of which are well-known enhanced ferromagnetic systems, show that the diffuse satellite peaks coexist with the ferromagnetic long-range order.[9] Polarized neutron measurements reveal that the transverse spin component of the ferromagnetic moment is modulated with a wave-vector incommensurate with the lattice periodicity and propagating along the [1 0 0] direction.[10] This is the same situation that is observed in the present PdMn system. Thus, the PdMn alloys with low Mn concentration may be considered to be the same type of enhanced ferromagnetic system as that found in the PdFe system. However, several difficulties exist for this model. In the PdFe system, the diffuse satellite peaks disappear together with the ferromagnetic long-range order at T_c,[9] while for the PdMn alloy, they survive up to four times the Curie temperature. Under weak magnetic fields, the diffuse satellite peak intensity should increase by 50% due to the realignment of ferromagnetic domains as observed in PdFe alloys,[9] but no such behavior was observed. The disappearance of the diffuse satellite peaks under the strong magnetic field may be explained as an alignment of the transverse spin component due to the magnetic field, but the increase in the background counts is not. The magnetization process which shows two step changes below T_c is well explained by this model, but it

is very difficult to explain the hard process above T_c.

(3) SDW cluster model: SDW clusters always have a uncancelled spin component when their correlation length is comparable with or shorter than their wavelength. The uncancelled spin component is very susceptible to the applied magnetic field and the ferromagnetic features result. If this case applies to the present system, a similar magnetic phase diagram is expected for PdCr (Ref. 11) and CuMn spin-glass alloys because in all these spin-glass alloys, dynamical fluctuation of the SDW clusters plays an essential role in the spin glass behavior. But only the PdMn alloy with low Mn concentration shows the ferromagnetic phase. We can explain this point as follows.

For PdCr alloys, the Kondo temperature is considered to be rather high (~100 K) and the Kondo singlet state is formed at low temperature. Thus, PdCr alloys with relevant Cr concentration (less than 7 at. % Cr) is nonmagnetic. It must be noted however that the mass susceptibility in the spin-glass region (more than 7 at. % Cr) increases with decreasing Cr concentration.[11]

In the case of CuMn alloys, the local magnetic structure is different from that in PdMn alloys since the diffuse satellite peaks are observed at the 1, $1\pm\eta$, 0 positions. In the plane perpendicular to the SDW propagation direction, the nearest-neighbor Mn spins couple antiparallel and the net moment in this plane within a SDW cluster would be small. That is to say, in the SDW clusters of CuMn alloys, the SDW modulation takes place between the antiferromagnetic planes.[7] On the other hand, since the diffuse satellite peaks in PdMn alloys are observed at the $1\pm\delta$,0,0 positions, the spins on the plane perpendicular to the SDW propagation direction couple parallel. Thus, the SDW modulation in PdMn alloys is between the ferromagnetic planes as shown in Fig. 9. The large uncancelled moments would be expected in the PdMn SDW clusters. This model, however, still includes a difficulty. It is rather hard to imagine how the SDW clusters collapse under the magnetic field. Hicks and Cable[12] studied the diffuse satellite peaks in CuMn spin-glass alloy under an applied magnetic field and observed a uniform reduction (~10%) of the magnetic scattering in a field of 4.25 T. They reported that the ferromagnetic and the antiferromagnetic correlations are intimately connected to the uniform magnetic response. In the PdMn case, however, the magnetic-field effect appears to be far more drastic. The PtMn alloy also shows a spin-glass-like behavior[13] and diffuse satellite peaks at the same symmetry positions are observed.[14] However, no ferromagnetic phase is reported for PtMn alloys.[13] We cannot explain the difference of the magnetic phase diagrams between the PdMn and PtMn alloys by this model. The drastic phase change from the ferromagnetic phase to the spin-glass phase as Mn concentration increases is also hard to explain using this model.

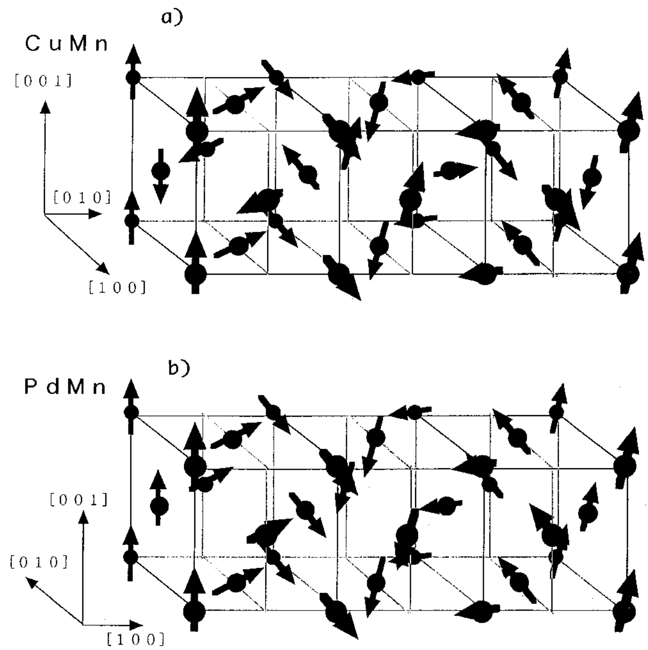

FIG. 9. SDW structure models for (a) CuMn and (b) PdMn alloys.

Among the models proposed here, the inhomogeneous cluster model seems to be the most plausible. However, this model does not give any idea of the origin of the ferromagnetic coupling. Although previous authors pointed out the importance of antiferromagnetic spin correlation for ferromagnetic PdMn alloys, their models with collinear antiferromagnetic coupling and isotropic spin configuration seem to be too simple to explain the incompatible features of this system. The present data suggest that the magnetic interaction depends on the direction. For instance, the SDW spin modulation in this alloy is a reflection of the special shape of the Fermi surfaces and propagates along the [1 0 0] direction because Pd metal has parallel plane Fermi (hole) surfaces perpendicular to the [1 0 0] axis. Thus, the spins in the plane perpendicular to the SDW propagation direction couple ferromagnetically [see Fig. 9(b)].

A more complete understanding of the magnetism of this system requires consideration of the whole band structure. Theoretical calculations are desired which take the actual band structure of the system into consideration.

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