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Oscillatory spin modulation in ferromagnetic disordered alloys $Pd_{1-y}Fe_y$ and $Pt_{1-x}Fe_x$

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Magnetic scattering of neutrons in the ferromagnetic disordered alloys $Pt_{1-x}Fe_x$ $(0.08 \leqslant x \leqslant 0.18)$ and $Pd_{1-y}Fe_y$ $(y = 0.1$ and $0.15)$ was measured using several single-crystal specimens. We found diffuse satellite reflections at $1 \pm \delta$, 0, 0 and the equivalent symmetry positions for both alloys except for PtFe alloys with $x > 0.13$. The wave vector of the spin modulation varies with Fe concentration. An incommensurate-commensurate transition takes place for the PtFe alloy at $x \sim 0.14$. The satellite reflections disappear at the Curie temperature of each specimen, suggesting that the oscillatory spin component homogeneously coexists with the ferromagnetic long-range order. An oscillatory spin polarization of band electrons due to the nesting of the Fermi surfaces is discussed as a possible origin of the satellite reflections. [S0163-1829(97)00817-5]

INTRODUCTION

Both Pd and Pt are well known as nearly ferromagnetic substances. An introduction of less than 1 at. % of Fe induces ferromagnetic long-range order (LRO) for PdFe. The phenomenon is interpreted by the induced spin polarization of Pd or Pt host atoms around Fe moments and named as a giant magnetic moment because the magnetic moment per Fe atom is as large as $10\mu_B$ for the PdFe alloy. Neutron-scattering measurements revealed that the spatial distribution of the spin polarization associated with the Fe impurities extends to distances exceeding 10 Å.[1] The Curie temperature for the ferromagnetic LRO increases with increasing Fe concentration for both systems.[2,3] Thus, PdFe and PtFe disordered alloys with an Fe concentration around 10 at. % are considered to be simple ferromagnets and only results from a few neutron-scattering experiments have been reported for these ferromagnetic alloys.[4,5]

On the other hand, more concentrated alloys, which show Cu_3Au-type chemical order, have been investigated intensively.[6–10] Bacon and Crangle studied the magnetic structure of a Pt_3Fe ordered alloy in detail[6] and reported that ordered Pt_3Fe shows antiferromagnetic LRO. However, ordered Pd_3Fe shows ferromagnetic LRO.[5]

In this paper, neutron-scattering data for disordered PtFe and PdFe alloys with Fe concentrations around 10 at. % are presented. For these alloys, we observed diffuse satellite reflections around the 1 0 0 reciprocal-lattice point (RLP). Since the satellite peaks disappear at the Curie temperature, these are magnetic in origin. Thus, we report here that both PtFe and PdFe disordered ferromagnetic alloys possess an antiferromagnetic spin correlation. The antiferromagnetic spin component is periodically modulated and the wave vector of the spin modulation varies with Fe concentration. For PtFe alloys, an incommensurate-commensurate transition takes place around 14 at. % Fe. The possible interpretation of the modulated antiferromagnetic component is given based on the oscillatory spin polarization of the band electrons.

SAMPLE PREPARATION AND MEASUREMENTS

Both Pd and Pt with a purity of 99.95% and Fe with 99.99% were used as raw materials for the present specimens. Various specimens were prepared for $Pt_{1-x}Fe_x$ with $x = 0.08$, 0.10, 0.12, 0.14, and 0.18 and for $Pd_{1-y}Fe_y$ with $y = 0.10$ and 0.15. Single crystals each with a volume of about 1 cc were grown by the Bridgman method using a furnace with a carbon heater system in an Ar atmosphere. Specimens were cooled down in the furnace and used for the neutron-scattering measurements in the as-grown state.

Neutron-scattering measurements were performed using a conventional-type triple-axis spectrometer T_{1-1} installed at a thermal guide of JRR-3M, Tokai. All of the data were taken with an incident neutron energy of 14 meV. A refrigerator unit was used for lowering the temperature of the specimens.

EXPERIMENTAL DATA

Diffraction patterns observed by scanning along the [100] direction on the (001) scattering plane are given in Fig. 1(a) for $Pt_{1-x}Fe_x$ alloys with various x and in Fig. 1(b) for $Pd_{1-y}Fe_y$ alloys with $y = 0.10$ and 0.15. These data were taken at the lowest temperature (=10 K). The sharp peak observed at 1 0 0 is contamination from the $\lambda/2$ (2 0 0) Bragg peak and indicates the exact position of the 1 0 0 RLP. Since broad peaks are observed on both sides of the 1 0 0, these are satellite reflections. The satellite peak positions shift with the Fe concentration and at $x = 0.14$, these peaks merge into a single peak at 1 0 0, indicating that the incommensurate-commensurate transition takes place around this composition for the PtFe system. The concentration dependence of the peak shift for PdFe is weaker than that for PtFe. The satellite peak linewidth along the direction perpendicular to the scattering vector is also studied and is almost the same as that along the [1 0 0] direction. The linewidth in the commensurate phase $(x \geqslant 0.14)$ is rather sharp and the intensity is far stronger.

An example of the temperature variation of the satellite peaks is given in Fig. 2 for a $Pt_{90}Fe_{10}$ alloy. The temperature dependences of satellite peak intensities for various PtFe alloys are plotted in Fig. 3. In this figure, the arrows indicate the reported values of Curie temperature[2] of the alloys with each Fe concentration. The satellite peaks disappear at the reported value of T_c. Temperature variation of the satellite peak position was also studied, but peak shift beyond the

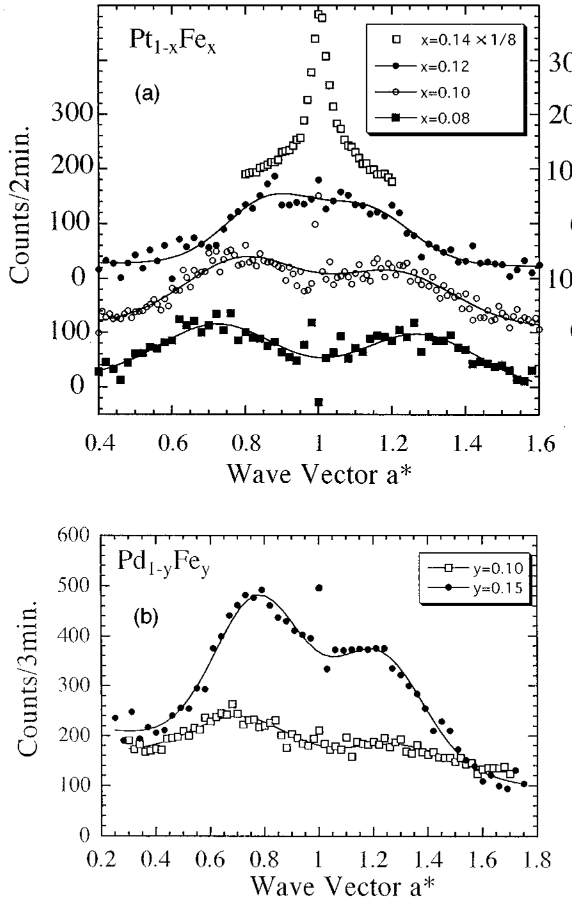

FIG. 1. (a) Satellite line profiles of various $Pt_{1-x}Fe_x$ alloys observed by scanning along the [1 0 0] direction. The high-temperature data were subtracted from the lowest-temperature (~10 K) data. The peak intensity for $x=0.14$ is very strong and the plotted values are multiplied by 1/8. Solid lines are best-fit curves using double Gaussians. (b) Experimental data for $Pd_{1-y}Fe_y$ alloys studied at 10 K. Raw data are plotted.

experimental error was not observed. Satellite reflections around other reciprocal-lattice points were also studied. Observed satellite peak positions in the (0 0 1) scattering plane are depicted in Fig. 4. It should be noted that these peak positions have the symmetry of an fcc lattice if we take the wave vector $\mathbf{Q}(=1-\delta)$ as a fundamental wave vector of the spin modulation, and are different from those of a Cu_3Au-type lattice which is well known for the Pt_3Fe ordered alloy.

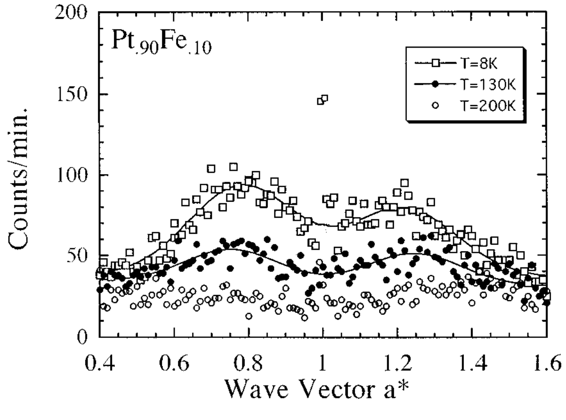

FIG. 2. Temperature variation of the satellite line profile for the $Pt_{.90}Fe_{.10}$ alloy.

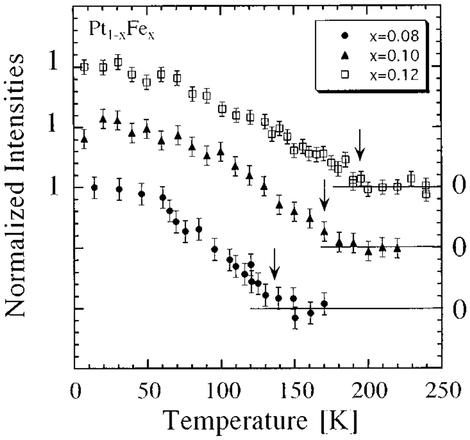

FIG. 3. Temperature variations of the satellite peak intensities studied for various PtFe alloys. The arrows indicate the reported Curie temperatures.

DISCUSSIONS

Prior to the neutron-scattering measurements, all of our specimens were confirmed to be ferromagnetic at 78 K using a permanent magnet. Nevertheless, we observed the satellite reflections for these specimens around the 100 RLP, indicating that the specimens have an antiferromagnetic spin correlation. Then, we can consider two possibilities; the antiferromagnetic component distributes homogeneously through the specimen (homogeneous model) or the specimens include mesoscopic scale inhomogeneities such as ASRO or concentration fluctuations. Experimental data, however, support the homogeneous model for the following reasons. (1) If the inhomogeneous model is valid, there must exist some concentration range for the ASRO to show the satellite reflections as being observed. However, there are no reports that PtFe and PdFe alloys show such satellite reflections in any concentration range or the ASRO. (2) The satellite peaks

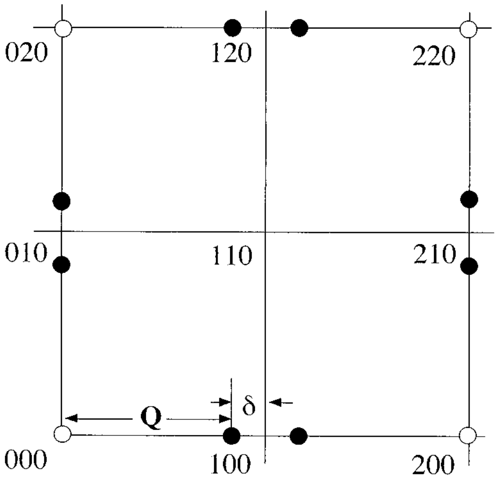

FIG. 4. Observed satellite peak positions in the (0 0 1) scattering plane.

disappear at the reported value of the Curie temperature at which the ferromagnetic LRO disappears. (3) The hyperfine fields for both PtFe and PdFe alloys with relevant Fe concentrations studied by the Mössbauer spectroscopy are well explained by a single hyperfine field.[2,3] (4) The satellite peak intensity depends strongly on the applied magnetic field. This suggests that the antiferromagnetic spin modulation accompanies the ferromagnetic LRO. Thus, the simple ferromagnetic structure is not valid for PtFe and PdFe disordered alloys. The antiferromagnetic correlation with incommensurate spin modulation coexists with the ferromagnetic moments.

Bacon and Crangle reported that an antiferromagnetic long-range order develops for the chemically ordered Pt_3Fe alloy which has a Cu_3Au structure.[6] The magnetic structure with antiferromagnetic coupling between neighboring Fe moments is reported for Pt_3Fe, in which Fe atoms occupy second-neighbor positions of the fcc lattice. Since the nearest-neighbor Fe spin favors a ferromagnetic coupling, frustration may be considered to be an origin of the modulated antiferromagnetic spin component in the disordered ferromagnetic alloys. However, this is not the case. The satellite peaks observed here are located around the 1 0 0 RLP, indicating that the antiferromagnetic coupling develops between the nearest-neighbor atoms of the fcc lattice and the ferromagnetic coupling is stabilized between the second-neighbor atoms. Thus, the modulated antiferromagnetic component has nothing to do with the antiferromagnetic LRO developed in the ordered Pt_3Fe alloy. Furthermore, we also observed satellite peaks at the same symmetry positions for the PdFe alloys, for which antiferromagnetic LRO has not been reported over the whole concentration range.

Since the satellite peak position shifts rather sensitively with the Fe concentration, the Fermi surface effect is a possible interpretation of the modulated antiferromagnetic component. Moriya[11] derived the expression for the spin polarization of alloys induced by magnetic impurity atoms. The magnetic moment of the jth atom is written as

$$m_j=\overline{m_j}+\alpha\sum{}' f(R_j-R_l)\overline{m_l},$$

where $\overline{m_j}$ is a spin polarization without considering the intra-atomic exchange interaction of the host metal and α is an enhancement factor due to the intra-atomic exchange. The function $f(R)$ is the normalized nonlocal susceptibility and is given as

$$f(R)=N^{-1}\sum \exp(ikR)\{f_0(k)/(1-\alpha f_0(k))\},$$

where $f_0(k)=\chi_0(k)/\chi_0$, $\chi_0(k)$ is a wave-number-dependent susceptibility and χ_0 is the uniform susceptibility without considering the intra-atomic exchange. At large R, $f(R)$ is approximately written as a screened Coulomb-type function, $f(R)\propto R^{-1}\exp(-\lambda R)$. Since λ tends to 0 as α comes close to unity, $f(R)$ has a singularity at k^{-2}. Thus, if α is very close to unity, this term overcomes the normal oscillatory term at moderately large R, where the latter comes from the logarithmic singularity at $2k_F$ in $f_0(k)$. This is accepted as an interpretation of the giant magnetic moments and strong ferromagnetic substances of PdFe and PtFe disordered alloys. The exchange enhancement factor $(1-\alpha)^{-1}$ is estimated to be 6~10 for Pd and 2~3 for Pt.[11]

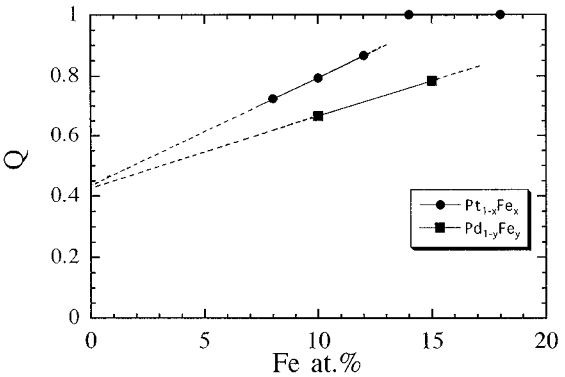

FIG. 5. Observed wave vectors of the oscillatory spin modulation plotted as functions of Fe concentration. Satellite peak positions were determined by the Gaussian fitting.

Although the enhanced susceptibility always shows a positive value due to the strong enhancement and the ferromagnetic coupling between Fe moments is realized for these alloys, the contribution of the oscillatory term may not be negligible, if the unenhanced susceptibility $\chi_0(k)$ is large enough. This situation is expected when the Fermi surfaces satisfy the nesting condition. The Fermi surfaces of Pd and Pt have almost similar shape[12] and we believe that these systems satisfy the nesting condition. In Fig. 5, the observed satellite peak positions, determined by Gaussian fits, are plotted as a function of the Fe concentration. Zero-point extrapolation of both the x and y values converge to almost the same point at $Q\sim 0.45$ (in 2π/a unit)$=0.72$ Å^{-1}. This value is not far from the distance of parallel planes of the parallel piped Fermi (hole) surfaces which are located at the W point and elongate along the cubic axes.[12] In connection with the present findings, we studied several PdM and PtM ($M=3d$ element) disordered alloys using neutron diffraction. For PdMn alloys, which are believed to be a spin-glass system, we observed strong satellite reflections at the same symmetry positions.[13] The satellite peak position depends on the Mn concentration. On the other hand, a PtV alloy shows temperature-independent satellite peaks at the same symmetry positions, indicating that chemical short-range order develops along the [1 0 0] direction.[14] These phenomena are considered to come from the same origin, the nesting Fermi surface effect in Pt and Pd alloys.

The above interpretation implies that the oscillatory spin component is parallel to the ferromagnetic spin polarization. In order to obtain knowledge about the actual spin structure, the satellite reflections are studied for $Pd_{.85}Fe_{.15}$ under a magnetic field at room temperature. When the magnetic field is applied along the direction parallel to the scattering vector, the satellite peaks completely disappear. When the magnetic field is applied perpendicular to the scattering plane, the satellite peak intensity increases by about 50%. The experimental data are given in Fig. 6. From this fact, we can discard some models of the spin structure, but are still unable to determine the actual spin structure definitely from the present experimental data alone. Further measurements using polarized neutrons are planned to study the actual spin structure of these systems.

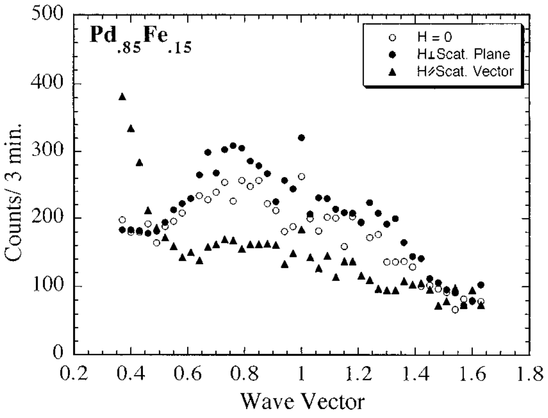

FIG. 6. Satellite peak line profiles for the $Pd_{.85}Fe_{.15}$ alloy studied in a magnetic field at room temperature. Open circles are the data without the magnetic field, triangles are with the field parallel to the scattering vector, and closed circles are with the field perpendicular to the scattering plane.

The line profiles of the satellite peaks are very broad. From the observed linewidth, the correlation length is estimated to be about 5 times the lattice parameter. Since the satellite peak position is very sensitive to the Fe concentration, one explanation of the broad line profiles is the inhomogeneous distribution of Fe concentration. However, the broadness seems to be an intrinsic feature of the random alloys. Even for the $Pt_{.88}Fe_{.12}$ alloy, fluctuations on a mesoscopic scale with 14 at. % Fe do not exist because no commensurate peak at 100 is observed. It is noted that the linewidth of the $Pt_{.86}Fe_{.14}$ alloy, which shows a commensurate antiferromagnetic peak at 1 0 0, is far sharper than that of the incommensurate satellite reflections observed for the alloys with $x<0.14$. In the case of incommensurate structures (helical or sinusoidal), there exists infinite freedom in the choice of the phases of the wave modulations. Thus, the correlation length becomes very short for the random alloys with incommensurate spin modulation. As noted above, Cable, Robertson, and one of the present authors found strong satellite reflections for PdMn alloys[13] at the same symmetry positions as those of PdFe. The linewidth of the satellite peaks is again broad. The spin correlation with incommensurate modulation in the disordered alloys cannot propagate to long range due to the infinite freedom of the phase relation. This is considered to be an essential point for the spin-glass-like behaviors of PdMn alloys and the situation is just the same as that in the typical spin-glass alloy CuMn.[15,16]

In addition to the 1 0 0 commensurate magnetic peak, another magnetic peak was observed at the 1/2 1/2 0 RLP for both $Pt_{.86}Fe_{.14}$ and $Pt_{.82}Fe_{.18}$ alloys. The peak intensity of the latter for $Pt_{.82}Fe_{.18}$ is far stronger than that for $Pt_{.86}Fe_{.14}$, while the 1 0 0 commensurate peak intensity decreases with increasing Fe concentration. Furthermore, temperature variations of the magnetic peak intensities for 1 0 0 and 1/2 1/2 0 are different. Bacon and Crangle reported that the Pt_3Fe ordered alloy has the antiferromagnetic structure for which the magnetic Bragg peak is observed at 1/2 1/2 0.[6] It is reasonable to conclude that the 1/2 1/2 0 magnetic peak comes from the Pt_3Fe-type ASRO.

Both PtCo and PdCo alloys are also known as strong enhanced ferromagnetic systems due to the same reasons as PtFe and PdFe systems. Measurements of satellite diffuse scattering for these systems are now in progress.

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