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LETTER TO THE EDITOR

Modulated spin structure of the enhanced ferromagnet $\mathbf{Pd_{85}Fe_{15}}$ determined using polarized and unpolarized neutron scattering

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Abstract. The magnetic structure of an exchange-enhanced ferromagnetic **Pd**Fe alloy was determined by polarized and unpolarized neutron scattering measurements. The polarized neutron data indicate that the magnetic moments are accompanied by an oscillatory spin component perpendicular to the ferromagnetic moments. Further measurements of unpolarized neutrons support the assertion that the oscillatory spin component can be described by a rotation wave.

PdFe disordered alloy is well known as a strongly enhanced ferromagnet. The introduction of even less than 1 at.% of Fe induces ferromagnetic long-range order (LRO) [1]. The Curie temperature increases with increasing Fe content [2]. Thus, **Pd**Fe disordered alloy with an Fe concentration of around 10 at.% is considered to be a simple ferromagnet. Very recently, however, two of the present authors (Tsunoda and Abe) found that the strongly enhanced ferromagnet **Pd**Fe with an Fe concentration of about 10 at.% shows diffuse satellite reflections around the 100 reciprocal-lattice points in unpolarized neutron scattering experiments, as shown in figure 1 [3]. The satellite peak position depends on the concentration of Fe. Thus, ferromagnetic **Pd**Fe is associated with an incommensurate spin modulation.

Since the satellite reflections are diffuse peaks, the correlation length of the spin modulation is rather short and is estimated to be only about three times the lattice parameter. This is probably due to the random distribution of Fe atoms. Similar diffuse satellite peaks were also observed for other enhanced spin systems: **Pt**Fe [3] and **Pd**Co [4]. To provide clues as to how to discuss the origin of such a phenomenon, it is useful to determine the actual spin structure. However, it is impossible to determine the actual spin structure of **Pd**Fe alloy from unpolarized neutron scattering data solely. The purpose of the present experiments is to determine definitively the actual spin structure of the strongly enhanced ferromagnet **Pd**Fe alloy using polarized and unpolarized neutron scattering.

In discussing the magnetic structure of this alloy, it is necessary to consider three fundamental vectors, $\boldsymbol{Q}$, $\boldsymbol{M}$ and $\boldsymbol{S}$, where $\boldsymbol{Q}$ is the wave propagation vector of the spin modulation, $\boldsymbol{M}$ is a ferromagnetic moment and $\boldsymbol{S}$ indicates the modulated spin component. From the previous neutron data, we know that the wave propagation vector $\boldsymbol{Q}$ is perpendicular to the modulated spin component $\boldsymbol{S}$ because the peaks are observed at the $1 \pm \delta, 0, 0$ reciprocal-lattice point where $\delta = 1 - |\boldsymbol{Q}|$. Furthermore, the satellite

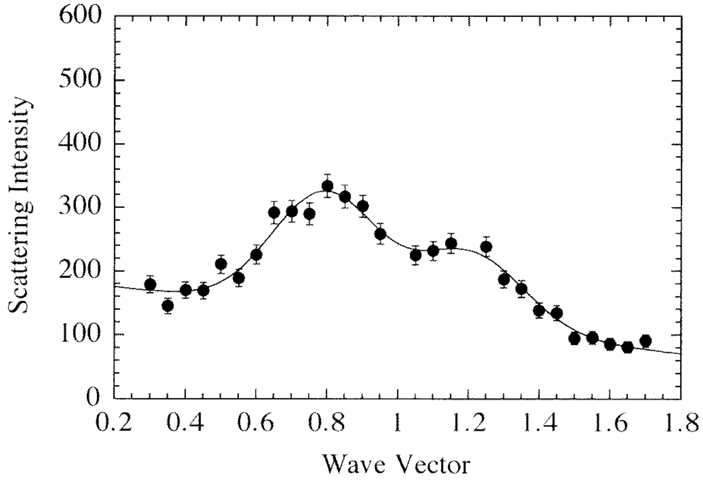

Figure 1. The satellite peak line profile observed by scanning along the [100] direction for $Pd_{85}Fe_{15}$ alloy using unpolarized neutron scattering at room temperature.

reflections disappear when the magnetic field is applied along the direction parallel to the scattering vector, indicating that one of the possible spin structures—that with $\boldsymbol{Q} \parallel \boldsymbol{M}$ and $\boldsymbol{S} \perp \boldsymbol{M}$—is not the one that we seek. Thus, our interest lies in determining which structure actually occurs in the **Pd**Fe alloy, (1) that with $\boldsymbol{Q} \perp \boldsymbol{M}$ and $\boldsymbol{S} \perp \boldsymbol{M}$ (figures 2(b) and 2(c)) or (2) that with $\boldsymbol{Q} \perp \boldsymbol{M}$ and $\boldsymbol{S} \parallel \boldsymbol{M}$ (figure 2(a)). These two possibilities may be distinguished by means of polarized neutron diffraction. However, in the former case, the magnetic structure is still not unique, because there are two possible choices for the modulated spin component. They are (1a) a sine wave (see figure 2(c)) and (1b) a rotation wave (a helix or frozen spin wave; see figure 2(b)) and may be distinguishable via unpolarized neutron scattering.

We now describe the sample preparation and measurements. A sample of nominal composition $Pt_{85}Fe_{15}$ was prepared from Pt with a purity of 99.95% and Fe with a purity of 99.99%. A single crystal with a volume of about 1.5 cm^3 was grown by the Bridgman method in an Ar atmosphere using a furnace with a carbon heater system. The specimen was cooled down in the furnace and used for the neutron scattering measurements in the as-grown state.

The polarized and unpolarized neutron scattering experiments were performed at the 5G (PONTA) and T_{1-1} triple-axis spectrometers, respectively, both installed at JRR-3M, Tokai. A Heusler alloy monochromator and analyser were used for the polarized neutron measurements.

The polarized neutron measurements were performed at room temperature (RT) under a vertical magnetic field of 3000 G and the scans were made along the [100] direction around the 100 reciprocal-lattice point. In these conditions, the ferromagnetic moment $\boldsymbol{M}$ is perpendicular to the scattering plane. Incoming neutrons polarized parallel to the magnetic field were used. Thus, spin-flip (SF) scattering detects the case in which the modulated spin component $\boldsymbol{S}$ is perpendicular to $\boldsymbol{M}$. On the other hand, non-spin-flip (NSF) scattering detects the case in which the modulated spin component $\boldsymbol{S}$ is parallel to $\boldsymbol{M}$. The flipping ratio in the present experimental conditions was about 7. This value is rather poor, but is still good enough for determining the qualitative features of the magnetic structure. The observed diffraction patterns for SF and NSF scattering are shown in figure 3.

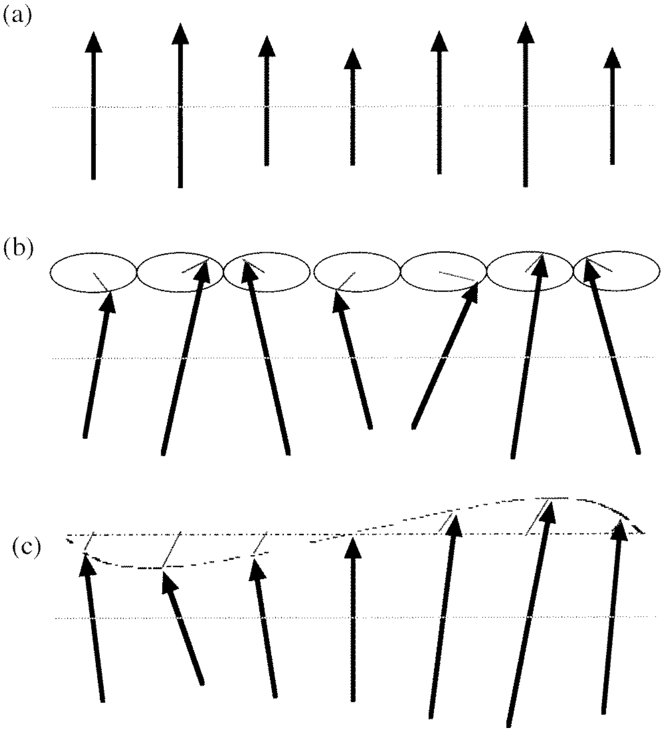

Figure 2. Illustrations of the modulated spin structures under consideration. (a) A longitudinal spin modulation for which the modulation component is parallel to the ferromagnetic moment. The transverse spin modulation is described as (b) a rotation wave or (c) a sine wave.

In SF scattering—see figure 3(a)—broad satellite peaks which have a similar line shape to the unpolarized neutron data (figure 1) are observed. In NSF scattering, however, there are no satellite-type reflections as shown in figure 3(b). NSF scattering includes contributions from nuclear scattering. Increasing of the intensities at low and high scattering angles in the scattering can be ascribed to the forward scattering and a skirt of the 200 Bragg reflections, respectively.

Thus, we can conclude that the modulated spin component $\boldsymbol{S}$ is perpendicular to the ferromagnetic moment $\boldsymbol{M}$. There still exist two types of modulation wave with $\boldsymbol{S} \perp \boldsymbol{M}$ as shown in figures 2(b) and 2(c). One is that for which the modulated spin component can be described as a sine wave. The other is the one for which it can be described as a rotation wave. It is possible to distinguish these two cases using unpolarized neutron measurements. Let us consider the satellite reflections in the $(1\bar{1}0)$ scattering plane. The satellite reflections appear at the $0, 0, 1\pm\delta$ and $1, 1, \pm\delta$ positions. Note that these satellite reflections come from the same magnetic domains. Since the spin component perpendicular to the scattering vector $\boldsymbol{K}$ is observable in magnetic neutron scattering, a half of the modulation spin component is observable for the rotation wave at the $0, 0, 1 \pm \delta$ satellite positions, but the full component is observable for the sine wave. On the other hand, at the $1, 1, \pm\delta$ satellite peak positions, three quarters of the rotation wave contributes to the magnetic scattering, but only a half of the sine wave does this. Thus, the intensity ratio $I(1, 1, \pm\delta)/I(0, 0, 1 \pm \delta)$ is 0.5 for the sine wave and 1.5 for the rotation wave, without taking the magnetic form factor into consideration. Assuming the magnetic form factor of Fe metal, these values must be

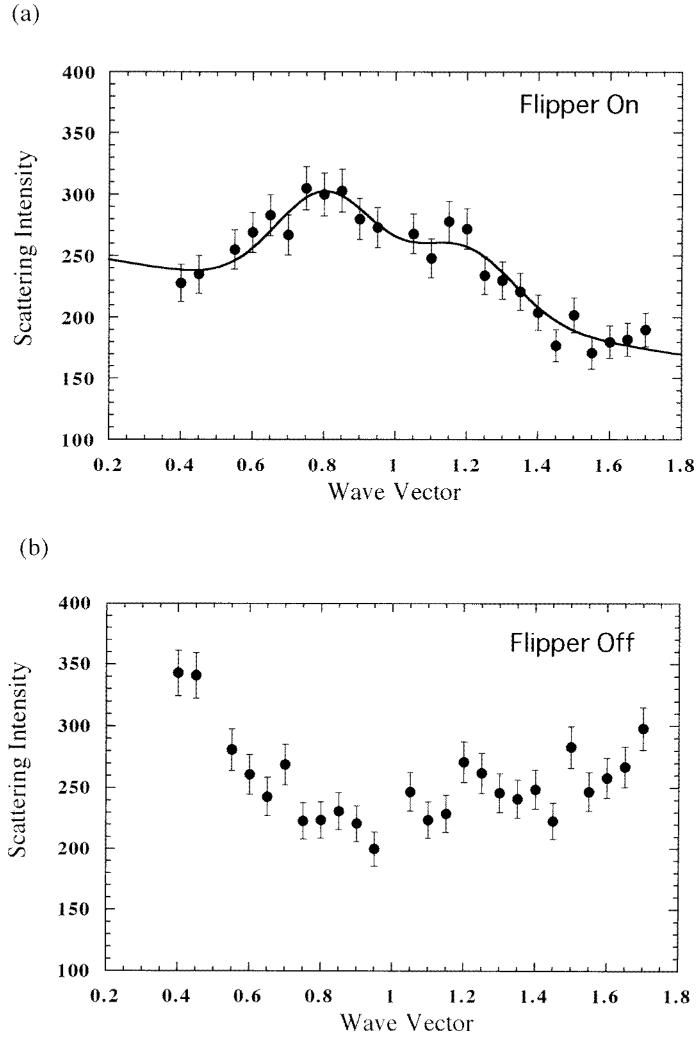

Figure 3. Polarized neutron scattering data taken under a vertical magnetic field. (a) Spin-flip scattering and (b) non-spin-flip scattering.

multiplied by the factor

$$\frac{|F(110)|^2}{|F(001)|^2} \approx 0.7.$$

The measurements described above were performed at 10 K and RT. The magnetic component was determined by the subtraction of these data; then we obtained the intensity ratio

$$\left.\frac{I(1,1,\pm\delta)}{I(0,0,1\pm\delta)}\right|_{ob} \approx 0.92.$$

This value is rather close to that for the case of the rotation wave, for which the intensity ratio is expected to be $1.5 \times 0.7 = 1.05$. Thus, the rotation wave (the frozen spin wave) is a better description for the **Pd**Fe ferromagnetic alloys. It must be noted that the reference value is estimated for the idealized spin system. The actual **Pd**Fe alloy would have a complicated spin structure with different sizes of magnetic moments.

It is obvious that the satellite peaks are observed on the both sides of the 100 reciprocal-lattice point only for SF scattering in the polarized neutron measurements. In order to confirm that these peaks are identical with those of the unpolarized neutron data, the polarized neutron data for the SF scattering were fitted using double Gaussians. The results were compared with unpolarized neutron data. From figure 3(a), the modulation wave vector δ ($\delta = 1 - |\boldsymbol{Q}|$), satellite linewidth and intensity ratio of the two satellite peaks are obtained as 0.19 ± 0.01 (in units of $2\pi/a$), 0.34 ± 0.04 (in units of $2\pi/a$) and 1.5 ± 0.2 respectively. The same analysis for the unpolarized neutron data shown in figure 1 gives us $0.21 \pm 0.01, 0.36 \pm 0.02$ and 1.6 ± 0.1, respectively. These values agree within the experimental errors.

Since the scattering amplitudes of Pt and Fe are nearly equal ($b_{\mathrm{Pt}} = 9.45$ b and $b_{\mathrm{Fe}} = 9.60$ b), it is impossible to observe the diffuse peaks arising from an atomic short-range order (ASRO) even if it exists. However, the ASRO in Pd–M and Pt–M alloys (M = 3d element) would be accompanied with lattice distortion. Such a system is clearly observed in PtV alloy for which the lattice spacing in the region with a high concentration of Pt expands [5]. Thus, if ASRO formed in the **Pd**Fe alloy, the asymmetric intensities of the satellite reflections would be expected due to the periodic lattice deformation. However, the observed intensity ratio $I(1-\delta, 0, 0)/I(1+\delta, 0, 0) \approx 1.5$ is explained by the magnetic form factor solely. Furthermore, the ASRO would form a longitudinal spin modulation due to the difference of the moment sizes of Fe and Pd, and indicate NSF scattering in the polarized neutron measurement. No appreciable diffuse satellite was observed in NSF scattering for the polarized neutron data. The amplitude of the ASRO would be rather small if it exists at all in the **Fe**Pd alloy.

Moriya derived an expression for the spin polarization for the strongly enhanced spin system induced by the magnetic impurity atoms [6]. The susceptibility always shows a positive value for the enhanced system, but the oscillatory term still persists. In this case, however, the theory suggests that the oscillatory part should be parallel to the ferromagnetic component, in contrast to the present experimental results. In the theory, general features of the giant moment are discussed. More realistic calculations based on the actual band structure are highly desired.

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