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# Anomalous magnetic moments in Fe–Pt and Fe–Pd Invar alloys under high pressure

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#### Abstract

Magnetization measurements have been carried out for disordered  $Fe_{72}Pt_{28}$ ,  $Fe_{66}Pd_{34}$ , and  $Fe_{68}Pd_{32}$  Invar alloys under high pressure using a technique combining a pressure-clamp-type Drickamer cell and a pulse magnet. In  $Fe_{72}Pt_{28}$  at room temperature, the magnetization decreased rapidly with increasing pressure up to 2.5 GPa, but above 2.5 GPa the rate of decrease became small and remained at a small value up to 5.6 GPa. In Fe–Pd Invar alloys at room temperature, the magnetization decreased linearly with increasing pressure. But, at 4.2 K, the change of magnetization with pressure was small in  $Fe_{66}Pd_{34}$ , which means that  $Fe_{66}Pd_{34}$  behaves as a strong ferromagnet.

### 1. Introduction

Binary and ternary alloy systems (e.g. Fe–Ni, Fe–Ni–Cr, Fe–Pt, Fe–Pd) in certain composition ranges show the Invar effect, i.e. thermal expansion is invariant at around room temperature [1]. In the case of Fe–Pt and Fe–Pd Invar alloys, the concentration dependence of the magnetic moment shows no deviation from the Slater–Pauling curve in the Invar range, which is quiet different from the case for the Fe–Ni Invar alloy. In addition, Fe–Pt Invar alloy behaves as a strong ferromagnet (FM) in contrast to the original Fe–Ni Invar alloy, which behaves as a weak FM [2].

Theoretical calculations for Invar alloys [3, 4] such as  $Fe_3Pt$  and  $Fe_{65}Ni_{35}$  showed the existence of two magnetic states that are energetically close: a high-spin (HS) state at larger volume with larger magnetic moment and a low-spin (LS) state at smaller volume with smaller magnetic moment. One can envisage a HS–LS transition taking place with increasing temperature or pressure. In particular, the calculation in [4] has predicted a pressure-induced HS–LS transition in Fe<sub>3</sub>Pt. Mössbauer spectroscopic (MS) measurements under high pressure [5] gave experimental evidence for this transition in disordered  $Fe_{72}Pt_{28}$ . They clarified that the hyperfine field ( $B_{eff}$ ) at 4.2 K decreases with increasing pressure up to 4.2 GPa

and the Curie temperature ( $T_C$ ) decreases to 40 K at 4.2 GPa, and then  $T_C$  and  $B_{eff}$  became constant with further increase of pressure. This was interpreted as showing that a pressureinduced phase transition from HS to LS took place in Fe<sub>72</sub>Pt<sub>28</sub> at 4.2 GPa and 40 K. On the other hand, x-ray magnetic circular dichroism (XMCD) measurements at room temperature [6] showed that a HS + LS state at ambient pressure transformed to a LS state at 4 GPa, and then transformed to a non-magnetic (NM) state at 20 GPa. The XMCD signal was observable up to 20 GPa at room temperature, which is in contradiction with the MS result. Therefore, we directly measured the pressure dependence of the magnetization in disordered Fe<sub>72</sub>Pt<sub>28</sub> Invar alloy by using an apparatus designed for complex extreme conditions of high pressure up to 5 GPa, pulsed magnetic field, and low temperature down to 4.2 K [7]. The magnetization decreased rapidly with increasing pressure, but a FM-like signal was observable throughout the pressure range at room temperature. However, the sensitivity of the measuring system was not sufficient to allow us to conclude that a FM-like magnetic phase definitely exists.

We have again carried out the magnetization measurements on  $Fe_{72}Pt_{28}$  under high pressure at room temperature, since we succeeded in improving the sensitivity of the measuring system; it now detects magnetization more clearly at high pressures around 5 GPa. Our measurements have been extended to Fe–Pd Invar alloys, too.

#### 2. Experimental details

The samples used in this study were disordered  $Fe_{72}Pt_{28}$ ,  $Fe_{66}Pd_{34}$ , and  $Fe_{68}Pd_{32}$  Invar alloys. Pressure was generated with a pressure-clamp-type Drickamer cell, which consists of a body made of Cu–Be alloy and zirconia, a pair of diamond anvils (1.5 mm diameter culet and 40° taper angle), and a pyrophyllite gasket [8]. The calibration of pressure was carried out by a ruby fluorescence technique using an optical fibre. The pulsed magnet used here can generate a maximum field of 20 T with the pulse width of 4 ms. The clamp cell was set at the centre of the bore of the magnet. The magnetization measurement was carried out by the system using a pick-up coil wound around the top of the anvil near the sample. The intensity of the magnetic field was calibrated by monitoring the cancelling coil wound around the cell.

## 3. Results and discussion

Figure 1 shows the observed dM/dt signals for Fe<sub>72</sub>Pt<sub>28</sub> at various pressures at 295 K, whereas figure 2 shows the signal without any sample obtained by using the same pick-up coil system. A ferromagnetic-like signal is still observable even at 5.6 GPa in figure 1.

Figure 3 shows the pressure dependence of the magnetization in  $Fe_{0.72}Pt_{0.28}$  at 295 K. The magnetization decreased rapidly with increasing pressure up to 2.5 GPa. This tendency is in good agreement with the result obtained by Hayashi and Mori [2]. Above 2.5 GPa the rate of decrease became small and tended to remain small up to 5.6 GPa. This was in contradiction with the previous MS result [5], in which  $T_C$  decreased to 295 K at around 2.5 GPa. However, the FM-like signals observed in the present experiment are in good agreement with XMCD measurements [6].

Fe–Pd alloys with the fcc phase around 30 at.% Pd show the Invar anomaly in the thermal expansion. On the other hand, a martensitic transition takes place in the concentration range of Pd < 34% at low temperature. We have carried out magnetization measurements on  $Fe_{66}Pd_{34}$  and  $Fe_{68}Pd_{32}$  at high pressure at 4.2 and 290 K, respectively.

Figure 4 shows the pressure dependence of the magnetization in Fe–Pd Invar alloys. At 290 K, the magnetization decreased linearly with increasing pressure in both  $Fe_{66}Pd_{34}$  and



**Figure 1.** Observed dM/dt curves for disordered Fe<sub>72</sub>Pt<sub>28</sub>. A: 0.1 MPa; B: 1.1 GPa; C: 2.5 GPa; D: 3.6 GPa; E: 5.6 GPa.

Fe<sub>68</sub>Pd<sub>32</sub>. The magnetization in Fe<sub>68</sub>Pd<sub>32</sub> decreased more rapidly with increasing pressure than that in Fe<sub>66</sub>Pd<sub>34</sub>, which is in good agreement with the magnetovolume effect in these alloys at room temperature. The pressure coefficients of magnetization (dM/dp)/M(0) are  $5.0 \times 10^{-2}$  GPa<sup>-1</sup> for Fe<sub>66</sub>Pd<sub>34</sub> and  $8.0 \times 10^{-2}$  GPa<sup>-1</sup> for Fe<sub>68</sub>Pd<sub>32</sub>. These values are of the same order of magnitude as that for Fe<sub>65</sub>Ni<sub>35</sub> at 295 K.

On the other hand, the magnetization of  $Fe_{66}Pd_{34}$  at 4.2 K decreased little. Fe–Pd Invar alloys does not show a large magnetovolume effect at low temperature. This result means that Fe–Pd Invar alloys behave as strong FMs at 4.2 K, which is the same behaviour as  $Fe_{72}Pt_{28}$  exhibits under pressures lower than 2 GPa, although  $Fe_{72}Pt_{28}$  above 2 GPa at 4.2 K shows a drastic decrease in magnetization as seen in the HS–LS transition observed in MS measurements [5] and the magnetization measurements [7]. These results indicate that no pressure-induced HS–LS transition took place in Fe–Pd Invar alloys in the observed pressure range.



Figure 2. The observed dM/dt curve without any sample.



Figure 3. The pressure dependence of the magnetization in  $Fe_{72}Pt_{28}$ .

# 4. Conclusions

In  $Fe_{72}Pt_{28}$  at room temperature, a small FM-like signal was observed at a high pressure of 5.6 GPa. This result indicates the possibility of the existence of a magnetic moment in  $Fe_{72}Pt_{28}$  under such high pressures.

The magnetization of Fe–Pd Invar alloys decreased linearly with pressure up to 5 GPa at 290 K. On the other hand, it does not decrease with increasing pressure at low temperature under high pressure up to 5.6 GPa. These results mean that the magnetovolume effect is very weak at low temperature. Fe–Pd Invar alloy behaves as a strong FM at low temperature. The magnetovolume effect in Fe–Pd Invar alloy is enhanced at elevated temperatures as seen for Fe–Pt Invar alloys.



**Figure 4.** The pressure dependence of the magnetization in Fe–Pd Invar alloys;  $Fe_{66}Pd_{34}$  at 4.2 K (O);  $Fe_{66}Pd_{34}$  at 290 K ( $\bullet$ );  $Fe_{68}Pd_{32}$  at 290 K ( $\triangle$ ).

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