

Study the Magnetic Properties of Invar Alloys by Using High Pressure Mössbauer Spectroscopy

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ABSTRACT

High pressure ^{57}Fe Mössbauer spectroscopy measurement (up to 42 Gpa) at room temperature have been carried out for investigation the magnetic properties of $\gamma(\text{f.c.c}) \text{Fe}_{78}\text{Ni}_{22}$ alloys using diamond anvil cell (DAC) technique. The Mössbauer spectrum at 0 Gpa shows a six line magnetic pattern with broad outer peaks and an average hyperfine field of $\sim 32\text{T}$ characteristic of a disordered alloys. In the pressure rang ($2 < P < 20$ Gpa) we observe Mössbauer spectra with additional low hyperfine field component resembling spectra of $\gamma(\text{f.c.c}) \text{Fe-Ni}$ Invar alloys (25-35 at % Ni). Our data indicate a pressure induced invar effect for $\text{Fe}_{78}\text{Ni}_{22}$ alloy at $\sim 7\text{-}12$ Gpa. Above 20 Gpa the hyperfine field break down and the alloy becomes non-magnetic showing only a single line Mössbauer spectrum.

Keywords – Mössbauer spectrum, $\text{Fe}_{78}\text{Ni}_{22}$, ^{57}Fe Mössbauer spectroscopy, ferromagnet

I. INTRODUCTION

Invar alloys are a very rich and interesting system. Particularly in the range between 22 and 50 at %, which has an f.c.c structure. In this region anomalous behavior is observed in some physical properties such as magnetostriction, thermal expansion etc [1]. Alloys which exhibit Invar behavior have a small thermal expansion coefficient below the Curie temperature, a large force volume magnetostriction and show a substantial pressure dependence of the magnetization and Curie temperature [2]. For alloys in the Fe-Ni system. Local fluctuations in compositions resulting in a heterogeneous magnetization are believed essential for the Invar effect.

Connection between the average magnetic moment and molar volumes is a common characteristic of $\gamma(\text{f.c.c}) \text{Fe-Ni}$ alloys, and ~ 30 at % Ni they show a near zero thermal expansion coefficient in a wide temperature range (the Invar effect). This effect and other related abnormal physical properties have been a subject of intense theoretical and experimental research over a century. Theoretical calculations predicted that the magnetic moment of Ni in $\gamma(\text{f.c.c}) \text{Fe-Ni}$ alloys appears to be nearly independent of the alloys composition or volume, whereas the moment of Fe change with volume [3]. Furthermore, the magnetic structure is characterized (even at zero temperature) by a continuous transition from a high moment ferromagnetic state at high volumes to a low moment disordered non-collinear configuration at low volumes [3].

^{57}Fe Mössbauer spectroscopy is a very valuable in high pressure research and sometimes gives unique

information regarding the magnetic properties of Iron containing materials under extreme pressure. Deportes, J. et al [4] studied the weak ferromagnets $\gamma(\text{f.c.c}) \text{Fe}_{68}\text{Ni}_{32}$ and $\text{Fe}_{65}\text{Ni}_{35}$ Invar alloys at variable temperature down to 2K and high pressure up to ~ 8 Gpa by ^{57}Fe Mössbauer spectroscopy using B4 Canvil. Their results showed that above a critical pressure e.g ~ 5.8 Gpa, the ferromagnetic state in $\text{Fe}_{68}\text{Ni}_{32}$ alloys is destroyed and the system displayed antiferromagnetic ordering at low temperature.

Here we study the strong ferromagnetic $\gamma(\text{f.c.c}) \text{Fe}_{78}\text{Ni}_{22}$ alloys by high pressure ^{57}Fe Mössbauer spectroscopy (up to ~ 42 Gpa) at room temperatures using diamond anvil cell (DAC) in order to investigate its magnetic properties through the pressure dependence of the Mössbauer hyperfine parameters.

II. EXPERIMENTAL

The alloy used in this investigation was prepared by are melting technique as described by [4]. The f.c.c structure of the alloy was confirmed by X-ray diffraction (lattice parameter $a=3.5804\text{\AA}$). The composition of the alloy was determined using microprobe analysis and its homogeneity was confirmed by SEM observations. High-pressure ^{57}Fe Mössbauer measurements were performed at room temperature in transmission geometry using DAC [5]. The spectrometer was calibrated using the spectrum of $\alpha\text{-Fe}$ at room temperature. NaCl was used as a pressure medium and several ruby chips were placed in the pressure chamber sealed with a Re gaskets, for pressure determination. Maximum

pressure gradient across the pressure chamber at 20 Gpa was less than 1 Gpa and at 40 Gpa less than 2 Gpa.

III. RESULT AND DISCUSSION

Figure 1 show some selected Mössbauer spectro of $^{57}\text{Fe}_{53}\text{Ni}_{47}$ alloy at different pressure collected on compression (a) and decompression (b). The spectrum at 0 Gpa is fitted with a magnetic sextet having an average hyperfine field at $31.4 \pm 0.5\text{T}$, characteristic of a high moment ferromagnetic alloy [6]. In the pressure range $2 < P < 20$ Gpa. We have fitted the Mössbaues field (solid and dashed sub spectra in Figure 1. respectively. For example at 8.5 Gpa. $B_H = 26.3\text{ T}$ and $B_L = 18.7\text{ T}$ see Figure 1(a). The line width and the Centre shifts were constrained to be equal for the two sextets during the fitting routine. We have also used the parameter dB%, which is a measure of the relative hyperfine field distribution with respect to the average field B, and the texture parameter T_p [7]. The spectra above 20 Gpa are fitted with broad singlets. In Figure 2(a) we show the pressure dependence of the normalized weighted average hyperfine field $[B(p)/B(0)]$ for the $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy. For comparison show on the same figure the data for $\text{Fe}_{68}\text{Ni}_{32}$ and $\text{Fe}_{65}\text{Ni}_{35}$ Invar alloys (at 4.2^ok) taken from reference [4]. As seen from Figure 2(a) and following the compression curve. $B(p)/B(0)$ which is generally proportional to the average magnetic moment of Fe, does not change with pressure up to ~ 2 Gpa. Above 2 Gpa it shows a gradual decrease with increasing pressure followed by a large negative pressure dependences similar to that observed in weak itinerant ferromagnets [4]. This implies that the ferromagnetic state in $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy has become unstable against pressure above ~ 2 Gpa, as a result of the decrease in the Curie temperature with increasing pressure. Our high pressure Mössbauer spectra of $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy in the range $2 < P < 20$ Gpa, which show an additional hyperfine field component, see Figure 1, resemble the Mössbauer spectra of Fe-Ni alloys in the Invar region ($30\% < \text{at} \% < 40$) at ambient conditions [6]. In Figure 2(b) we show for comparison the Mössbauer spectrum of the $\text{Fe}_{78}\text{Ni}_{22}$ alloy collected on compression at 7-6 Gpa (lower) and that of a commercial Invar $\text{Fe}_{64}\text{Ni}_{36}$ alloy measured at ambient pressure (upper). The two spectra are essentially identical and the hyperfine fields B_H and B_L are equivalent with in errors. This is in excellent with high-pressure X-ray diffraction studies on f.c.c

Fe-Ni alloys, where a pressure induced Invar effects has been observed at 7.7 Gpa (around RT) for $\text{Fe}_{55}\text{Ni}_{45}$ alloy [4]. On decompression we observe a typical Invar spectrum at 12.6 Gpa (see Figure 1(b), which indicates a pressure hysteresis as evident from Figure 2(a). This hysteresis is also seen in the pressure dependence of the Mössbauer relative intensity of the low field component as shown in Figure 3(a), a feature that has been reported in $\alpha \rightarrow \epsilon$ transition in pure Fe [8].

The pressure dependence of the Centre shift of $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy is shown in figure 3(b). Note the decrease in the CS with increasing pressure as a result of volume decrease. That is very close to the CS's for mechanically alloyed γ (f.c.c) Fe-rich Fe-Ni alloys.

Obtained at ambient conditions, where an antiferromagnetic ordering (with a low moment) indicated by the dramatic line broadening of the Mössbauer singlet was found to occur at $\sim 40\text{k}$ for γ (f.c.c) $\text{Fe}_{76}\text{Ni}_{24}$ alloy [3].

Suggested a pressure induced anti-ferromagnetism in γ (f.c.c) $\text{Fe}_{68}\text{Ni}_{32}$ alloy at 7 Gpa with TN of $\sim 35\text{k}$. in agreement with the local magnetic moment model [9], where a pressure induced increase of "latent" antiferromagnetism leading to an antiferromagnetic ordering at sudden decrease in the CS at ~ 20 Gpa may indicate a high moment to low moment transition. Above 20 Gpa, the CS's are fitted smoothly to a straight line that has an intercept of -0.07 mm/s at 0 Gpa. This value high pressure is expected. It appears from the above discussion that as pressure increases and volume decreases we observe a continuous transition from a stable ferromagnetic state with a high moment to a less stable one and eventually to a low magnetic moment above 20 Gpa. This is in agreement with theory [2] and it demonstrates the dependence of the magnetic moment on volume for γ (f.c.c) Fe-Ni alloys. It would be interesting to investigate the magnetic nature of the high pressure phase (above 20 Gpa), and a suitable method is to conduct high pressure ^{57}Fe Mössbauer experiments at low temperatures and in external magnetic field.

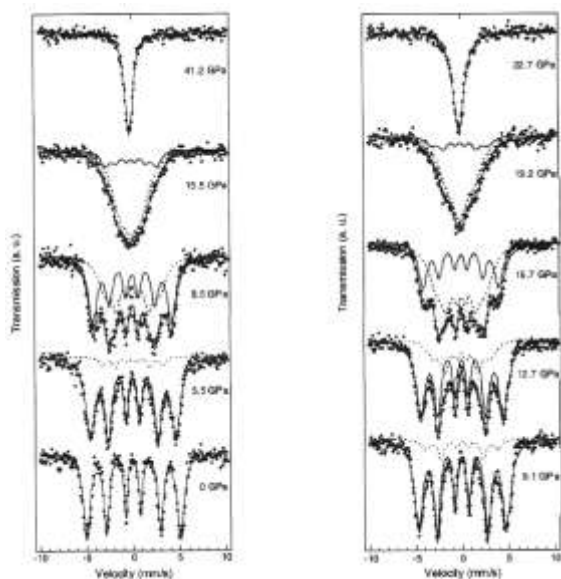


Figure 1: High-pressure Mössbauer spectra of $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy collected at RT: (a) compression: (b) decompression.

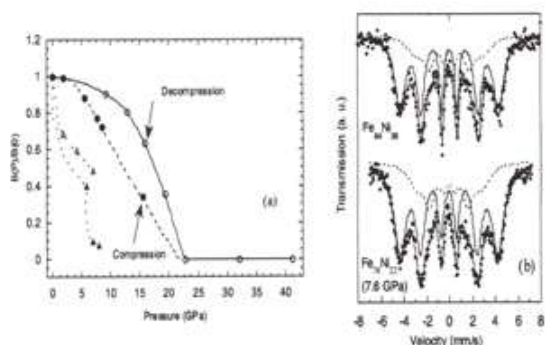


Figure 2. (a) Pressure dependence of the normalized weighted average hyperfine for $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy at RT: filled circles (compression), open circles (decompression). Open and filled triangles represent data from [3] (at 4.2 K) for Invar Fe65Ni35 and Fe69 Ni32, respectively. The lines through the data points are only guides to the eye. (b) RT Mössbauer spectra for $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy at 7.6 GPa (lower) and Fe64Ni36 Invar alloy at ambient pressure (upper).

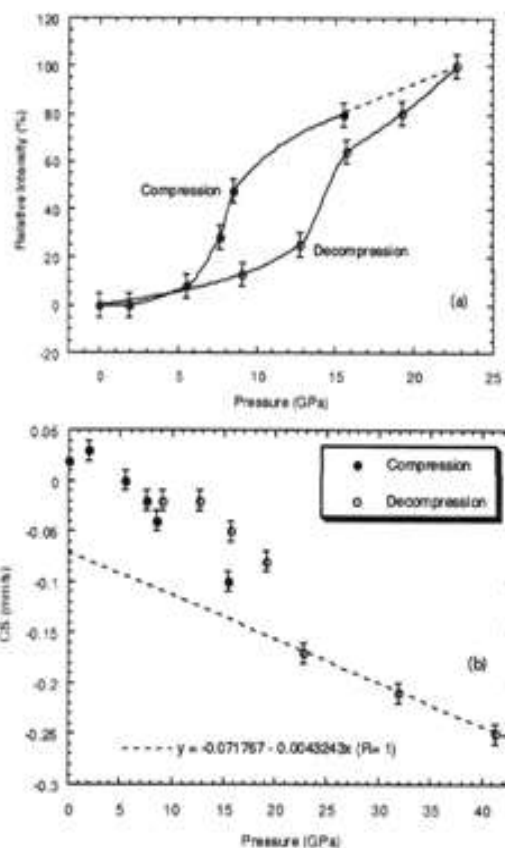


Figure 3. Pressure dependence of: (a) relative intensity of the low field component; (b) Mössbauer centre shift for $^{57}\text{Fe}_{78}\text{Ni}_{22}$ alloy. The dashed line in Figure 3(b) is linear fit to the CS's above 20 GPa.

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