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LETTER TO THE EDITOR

A ^{57}Fe high-pressure Mössbauer study of the ferromagnetic γ' - Fe_4N

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Abstract. The effect of pressure up to 5.6 GPa on the magnetic behaviour of γ' - Fe_4N has been investigated using the ^{57}Fe high-pressure Mössbauer effect technique at 300 K. We show that the decrease of the average magnetic hyperfine field at 300 K in this pressure range results from the decreases of the Fe local magnetic moment and Curie temperature with pressure. The decreases of the average isomer shift and the isomer shift for each Fe site with increasing pressure indicates a corresponding increase of s electron density at the ^{57}Fe nucleus, which is mainly caused by the volume compression of the 4s conduction electrons and changes of charge transfer between atoms.

In the last several years the interstitial rare earth–iron compounds have attracted much interest [1–6]. The interstitial atoms cause an expansion of the unit cell volume (e.g. about 6–7% for $\text{R}_2\text{Fe}_{17}\text{N}_{3-\delta}$) [6], and thereby give rise to a remarkable improvement of magnetic properties. Therefore, the study of these compounds under high pressure [7]—by which the volume of the unit cell can be reduced—is very helpful in obtaining a deeper understanding of the influence of the interstitial atoms on the magnetic properties. γ' - Fe_4N [8, 9] with cubic symmetry can be understood as a volume-expanded γ -Fe with an insertion of an N atom at the octahedral interstitial site. The simplicity of the crystal structure makes γ' - Fe_4N more suitable for the study of the effect of the interstitial atoms on the magnetic properties.

In this work, we have used the ^{57}Fe high-pressure Mössbauer effect (ME) technique (up to 5.6 GPa and at 300 K) in order to investigate the effect of pressure on the magnetic and electronic properties of the γ' - Fe_4N compound. The ^{57}Fe high-pressure ME spectroscopy offers us the possibility of investigating pressure-induced changes of the magnetic properties (via the effective magnetic hyperfine (HF) field at the ^{57}Fe nuclei, B_{eff}) and of the electronic properties (via the ME isomer shift, IS).

The powder of γ' - Fe_4N was prepared by heating directly ferrous oxalate, which was obtained by the chemical coprecipitation method [10], in a mixed stream of ammonia and hydrogen (a ratio of 1:1) in a quartz tube furnace at 550 °C for 1 hour. The x-ray diffraction pattern obtained by using Cu $K\alpha$ radiation shows a single phase of γ' - Fe_4N . The ^{57}Fe high-pressure ME experiments were performed in a Chester–Jones-type high-pressure set-up with B_4C anvils. The sample in the high-pressure cell was mixed with epoxy and placed in a pyrophyllite ring supported from outside by a steel belt. This type of pressure cell allows quasihydrostatic pressures up to 7.0 GPa to be attained. The pressure calibration at room temperature was performed by measuring the Mössbauer isomer shift of a β -Sn foil, whose pressure dependence has been calibrated [11, 12]. The B_4C anvils contain ^{57}Fe impurities that cannot be removed without destroying the mechanical properties of this material. Because of the presence of these impurities, there is a complicated ME spectrum

superimposed on the ME spectrum of the sample studied. For this reason, all the spectra were obtained by subtracting the spectrum of the anvils from those collected at each pressure point.

There are two non-equivalent crystallographic sites of the iron atoms in the γ' -Fe₄N structure, which are the cubic corner (Fe^I) and the face-centred (Fe^{II}) sites. Because the easy magnetization of γ' -Fe₄N is parallel to the [100] direction, the Mössbauer subspectrum for the Fe^{II} site is split into an additional subspectrum with an intensity ratio of 1:2. Therefore the Mössbauer spectrum of the γ' -Fe₄N was fitted to three sextets with an intensity Fe^I:Fe^{II}₁:Fe^{II}₂ = 1:1:2 [9]. Some selected Mössbauer spectra at different pressure points are shown in figure 1. We have deduced the average values of B_{eff} , $\overline{B}_{\text{eff}}$, from the weighted average values of B_{eff} of each subspectrum. The same analysis has been used to calculate the average values of the isomer shift ($\overline{\text{IS}}$).

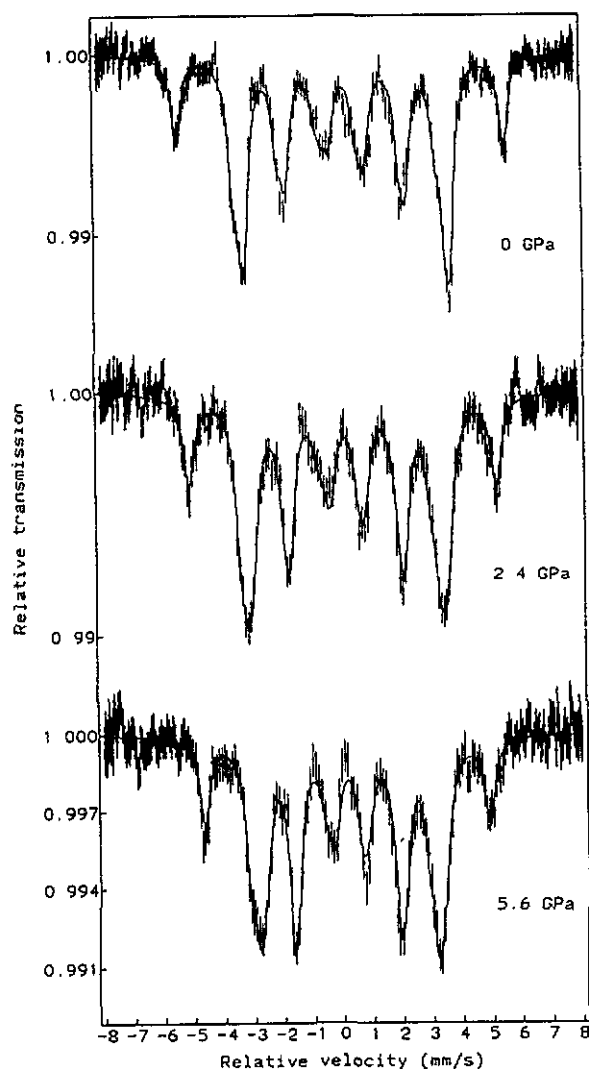


Figure 1. ⁵⁷Fe Mössbauer spectra of γ' -Fe₄N at 300 K and at pressures of 0, 2.4, 5.6 GPa.

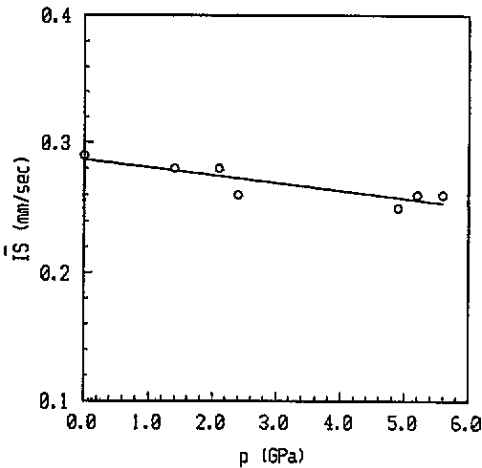


Figure 2. The pressure dependence of the average isomer shift \bar{IS} in γ' -Fe₄N at 300 K. The solid line through the data points is only a guide to the eye.

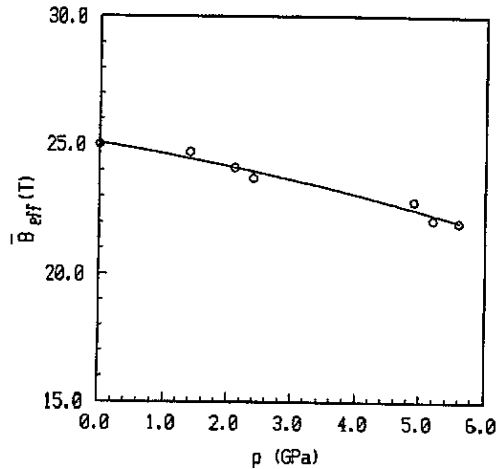


Figure 3. The pressure dependence of the average magnetic hyperfine field \bar{B}_{eff} in γ' -Fe₄N at 300 K. The solid line through the data points is only a guide to the eye.

The average values of \bar{IS} and \bar{B}_{eff} at room temperature as a function of pressure are shown in figure 2 and figure 3, respectively. It can be seen that both \bar{IS} and \bar{B}_{eff} decrease with increasing pressure. The decrease of \bar{IS} due to increasing pressure results from the increase of the s electron density at the ^{57}Fe nucleus $\rho(0)$ [13]. The linear pressure-induced decrease of isomer shift can be fitted well by using the formula

$$\bar{IS}(p) = \bar{IS}(0) - ap$$

where a is the slope, $a = \partial\bar{IS}/\partial p \simeq 5.9 \times 10^{-3} \text{ mm s}^{-1} \text{ GPa}^{-1}$ and $\bar{IS}(0)$ at $p = 0 \text{ GPa}$, $\bar{IS}(0) = 0.29 \text{ mm s}^{-1}$. Different mechanisms can contribute to an increase of $\rho(0)$ with increasing pressure. Using a value of the compressibility (κ) of about $5.1 \times 10^{-3} \text{ GPa}^{-1}$ for γ' -Fe₄N [14], one obtains $\partial\bar{IS}/\partial \ln V \simeq 1.2 \text{ mm s}^{-1}$, where V is the volume. This value is comparable with that for α -Fe at 300 K ($\partial\bar{IS}/\partial \ln V = 1.4 \text{ mm s}^{-1}$ [15]). Similarly we can fit the pressure-induced decrease of the isomer shift IS for Fe^I and Fe^{II} sites by using the above formula, respectively. The fitting results showed that the pressure-induced decrease of IS for an Fe^{II} site with a slope $\partial IS_{\text{Fe}^{\text{II}}}/\partial p \simeq 7.6 \times 10^{-3} \text{ mm s}^{-1} \text{ GPa}^{-1}$ and $\partial IS_{\text{Fe}^{\text{II}}}/\partial \ln V \simeq 1.5 \text{ mm s}^{-1}$ is more remarkable than that for an Fe^I site with a slope $\partial IS_{\text{Fe}^{\text{I}}}/\partial p \simeq 1.4 \times 10^{-3} \text{ mm s}^{-1} \text{ GPa}^{-1}$ and $\partial IS_{\text{Fe}^{\text{I}}}/\partial \ln V \simeq 0.3 \text{ mm s}^{-1}$. We can therefore suggest that the volume compression of the 4s conduction electrons, which results in the increase of $\rho(0)$, and the changes of charge transfer [16] ($\text{Fe}^{\text{I}} \rightarrow \text{Fe}^{\text{II}}$, $\text{N} \rightarrow \text{Fe}^{\text{II}}$), which increase with pressure and result in a decrease of $\rho(0)$ for the Fe^I site and an increase of that for Fe^{II}, are of comparable importance.

The decrease of \bar{B}_{eff} caused by decreasing the unit cell volume of γ' -Fe₄N under high pressure and at 300 K is evident from figure 3. Since the value of the Curie temperature T_C for γ' -Fe₄N at ambient pressure ($T_C \sim 760 \text{ K}$ [17]) is far above room temperature, it is suggested that the measured values of B_{eff} at room temperature and at different pressures will be affected simultaneously by the decrease of T_C and the Fe local magnetic moment with pressure. If it is assumed that the reduction of B_{eff} with increasing pressure is

mainly caused by the corresponding reduction of T_C , we can estimate the required reduction of T_C at the maximum pressure (5.6 GPa) which gives the experimental value of \bar{B}_{eff} (at 5.6 GPa) at 300 K by using the known temperature dependence of the magnetization (Brillouin curve for $J = \frac{7}{2}$). We then obtain a value of $T_C = 517$ K at 5.6 GPa which leads to a value of $\partial T_C / \partial p = -45$ K GPa $^{-1}$. This value of $\partial T_C / \partial p$ is considerable higher than the experimentally measured values of $\partial T_C / \partial p$ in $R_2\text{Fe}_{17}$ compounds [18]. Furthermore the ^{57}Fe high-pressure ME experiments at 4.2 K indicate a decrease of \bar{B}_{eff} with a value of $\partial \bar{B}_{\text{eff}} / \partial p = -0.26$ T GPa $^{-1}$ [14], which is caused by the decrease of the Fe local magnetic moment. Therefore it can be noted that the influence of the changes of Fe local magnetic moment are of importance and should be accounted. A detailed study of this last point will be published in a forthcoming paper [19].

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