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MÖSSBAUER STUDY OF AMORPHOUS FeF_3 , 0.4 HF IN THE MAGNETIC TRANSITION RANGE

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Abstract. – Analysis of Mössbauer spectra by unconstrained or constrained shaped distributions of both quadrupolar splitting and hyperfine field in the magnetic transition range centered on 39 K shows that spectra can be interpreted from static broadened field distribution only up to 36 K.

Recently, amorphous FeF_3 , $x\text{HF}$ ($0 < x < 1$) were synthesized by a soft chemistry reaction and studied by Greneche *et al.* in the range 4.2-400 K [1, 2]. This compound, speromagnetic at low temperature, reveals a magnetic behaviour similar to that of a spin glass with a maximum susceptibility cusp at $T_g=26$ K and a broad magnetic freezing centered near $T_m = 36$ K evidenced by Mössbauer thermal scanning [2].

The present Mössbauer study is focused on the spin freezing (temperature range 30-50 K with regulation better than 0.5 K). Zero field Mössbauer spectra give evidence of both paramagnetic and magnetic contributions. Analysis of spectra is complicated because of the amorphous structure of FeF_3 , which implies both distribution of quadrupolar splitting $P(\Delta)$ and hyperfine field $P(H)$ in the transition temperature range. These two distributions have to be separately extracted from the spectra. Spectra were first analysed by a sum of discrete quadrupolar and hyperfine field static distributions with unconstrained shape (free distributions). In spite of a satisfactory fitting, the overlapping part (in mm/s) of the two distributions can not be shared unambiguously between $P(\Delta)$ and $P(H)$, leading to some unphysical oscillations for the low field part of $P(H)$. To overcome this difficulty, spectra were fitted by constraining $P(H)$ to be asymmetric Gaussian shaped. For this distribution, maximum value of H , initial value of $P(H)$ and parameter of asymmetry are fitted parameters [3]. This method was powerful for spectra between 30 and 36 K where $P(H)$ is extended towards low fields. Above 36 K, where $P(H)$ is extended towards high fields, this method did not work well due to the chosen analytical distribution which is not well adapted to extension towards high fields and unconstrained distributions were used. Some fitted spectra are in figure 1. The isomer shift does not show any discontinuity. The paramagnetic fraction curve (Fig. 2) confirms a broad transition in the range 30-50 K with a transition temperature $T_M = 39$ K (defined for $\text{PF} = 0.5$) slightly higher than the previous result (36 K) obtained by thermal scanning on a different sample. The field distributions (Fig. 3) give ev-

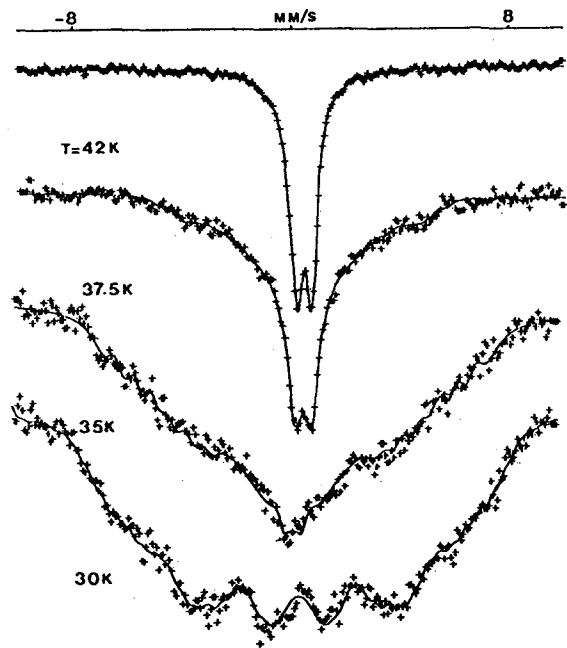


Fig. 1. – Spectra in the temperature range 30-42 K.

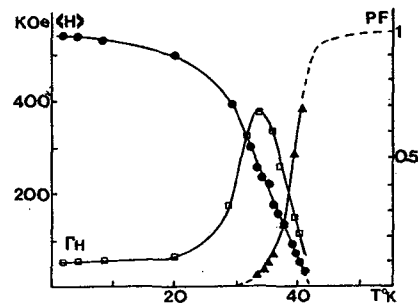


Fig. 2. – Thermal variation of average hyperfine field $\langle H \rangle$, width of the field distribution Γ_H and paramagnetic fraction PF.

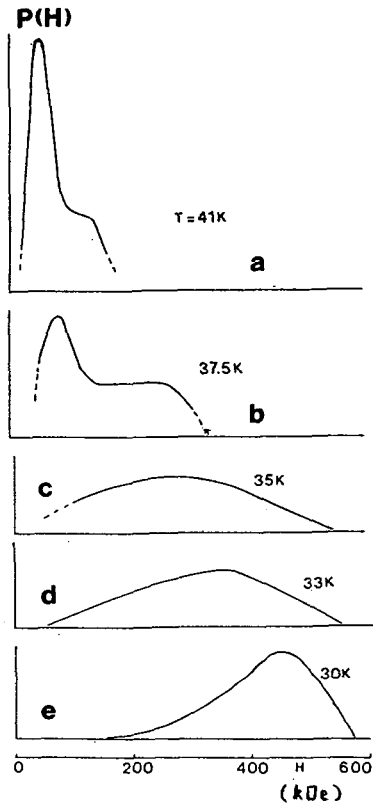


Fig. 3. - Fitted hyperfine field distributions (a, b: unconstrained shape - c, d, e: constrained shape).

idence for the changing of the asymmetry in the shape about 36 K. The average magnetic field $\langle H_n \rangle$ is reported in figure 2 together with Γ_H the width at half height of $P(H_n)$. The maximum of Γ_H is located at 35 K = 0.875 T_M . For high temperatures, the shape of the field distribution is unlikely, perhaps indicating that analysis of spectra requires correlated $P(\Delta)$ and $P(H)$ joint distributions. The results seem to be very similar to those on Au-3 % at Fe [4] and Rh-5 % at Fe [5] metallic spin-glasses. A static field distribution provides a satisfactory fit up to 36 K, but do not excludes dynamical models. At higher temperatures, relaxation effects should be taken into account.

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