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⁵⁷Fe HYPERFINE INTERACTION MEASUREMENTS IN AN IRON CONCENTRATED OXIDE GLASS Fe₂O₃(30)BaO(45)B₂O₃(25)

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We have investigated the preparative, structural, electronic and magnetic properties of an iron concentrated oxide glass Fe₂O₃(30)BaO(45)B₂O₃(25) and of its recrystallisation products. The present report concentrates on the measurement of the ⁵⁷Fe hyperfine interaction parameters in the amorphous phase (between 1.5 and about 680 K) and on their interpretation in terms of distributions of these parameters.

The glass is prepared by a splat cooling technique. Baryum borate and Fe₂O₃ are heated in nominal proportions at 1050 K for 6 hours, melted in a special crucible and quenched between steel rollers [1]. Thin ribbons are thus obtained. Their amorphous nature is characterized from X-ray diffraction and electron microscopy. Crystallisation involving the formation of successive phase is observed from DTA, X-ray diffraction and Mössbauer spectroscopy above ~ 700 K.

In the temperature range from 39 to 680 K, the Mössbauer spectra present a symmetric quadrupole doublet, with resonance widths (FWHM) estimated from a sum of two Lorentzians fit of 0.78 and 0.53 mm/s at these respective temperatures. The doublet develops into a pure magnetic hyperfine spectrum between 40 and 30 K ; in this temperature range one observes unresolved spectra, comporting apparently a superposition of the high temperature doublet and of a magnetic component with increasing relative intensity. The magnetic hyperfine spectra are fully symmetric around their center

of gravity below 29 K, in apparent contradiction with the high temperature quadrupole interaction. However, the linewidths of the spectral components are unequal ; sum of Lorentzian fits provides FWHM of 1.50, 1.08 and 0.75 mm/s for the outer, middle and inner components respectively at 4.2 K. The relative intensities of these components, extrapolated to zero thickness absorbers, differ from the theoretical ratio of 3 : 2 : 1 whereas the relative component areas fit satisfactorily with these values.

Several models have been applied to analyze these results in terms of distributions of hyperfine interactions. A RHSP model predicts a distribution of quadrupole splitting

$$(\Delta = 1/2 e^2 q Q (1 + \eta^2/3)^{1/2}) \text{ given by [2] :}$$

$$P(\Delta) = \Delta^4 \exp - (\Delta^2/2\sigma^2)$$

This model has been tested to fit the quadrupole data. However, better fits are obtained with a pure Gaussian distribution of Δ around an average value Δ_0 and a FWHM for the distribution WG as summarized in Table 1 (fig. 1). Of course, the respective distributions of V_{zz} and η cannot be separated out. There is no indication for any correlation between isomer shift and the distribution in Δ .

Table 1

Temperature (K)	673	300	45
Δ_0 (mm/s)	0.884	1.00	1.05
WG (mm/s)	0.79	0.94	1.00
δ_{IS} (mm/s) vs Fe - 300K	-0.015	0.262	0.363

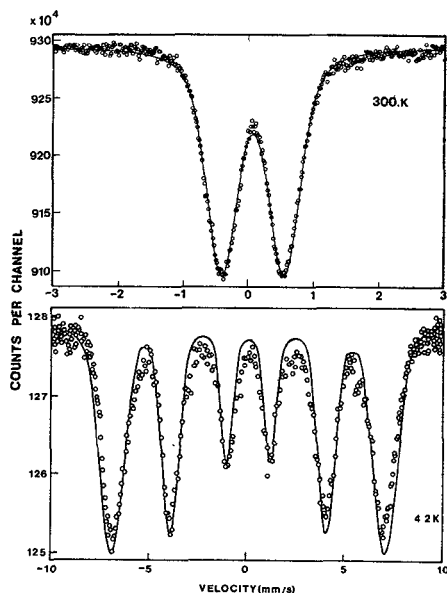


Fig. 1 : ^{57}Fe Mössbauer spectra at 300 and 4.2 K. The quadrupole spectrum is fitted with a gaussian distribution of Δ . The solid line in the 4.2 K spectrum is a simulation based on the approximations discussed in the text and with spectral component intensities constrained to relative values of 3 : 2 : 1.

The symmetry of the low temperature magnetic spectra point out that the distribution of Δ is symmetric about zero and is around both $+\Delta_0$ and $-\Delta_0$. The primary features of the magnetic spectra are reproduced by the following model calculation (fig. 1): a) The electric field gradient is assumed of axial symmetry ($\eta = 0$), b) The distribution of V_{zz} is constrained to the Gaussian distribution determined for Δ at 42 K around positive and negative Δ_0 , c) A spherical orientation between V_{zz} and hyperfine field axis and d) An additional Gaussian distribution of hyperfine field around an average value of 433 kOe and a FWHM of

50 kOe. This suggests exchange interactions of random character and dominating the anisotropy interactions, which is a reasonable conclusion for an S state ion in an amorphous oxide. Thickness and orientational effects have been shown experimentally to be negligible in the magnetic spectra. The deviation between calculated and experimental data in Fig.1 is thought to arise from the debatable assumptions of zero asymmetry parameter and spherical distribution of θ .

The nature of ordered structure, of magnetic transition and of recrystallisation behavior will be discussed elsewhere. Structural model calculations interpreting the quadrupole interaction distribution are known to be difficult in a random network model appropriate for such oxide glasses, where every information indicates that iron participates in the structure as a network former. It will be worthwhile to check for the validity of the observed distribution in other comparable systems.

References

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- /2/ Czjzek, G., private communication.