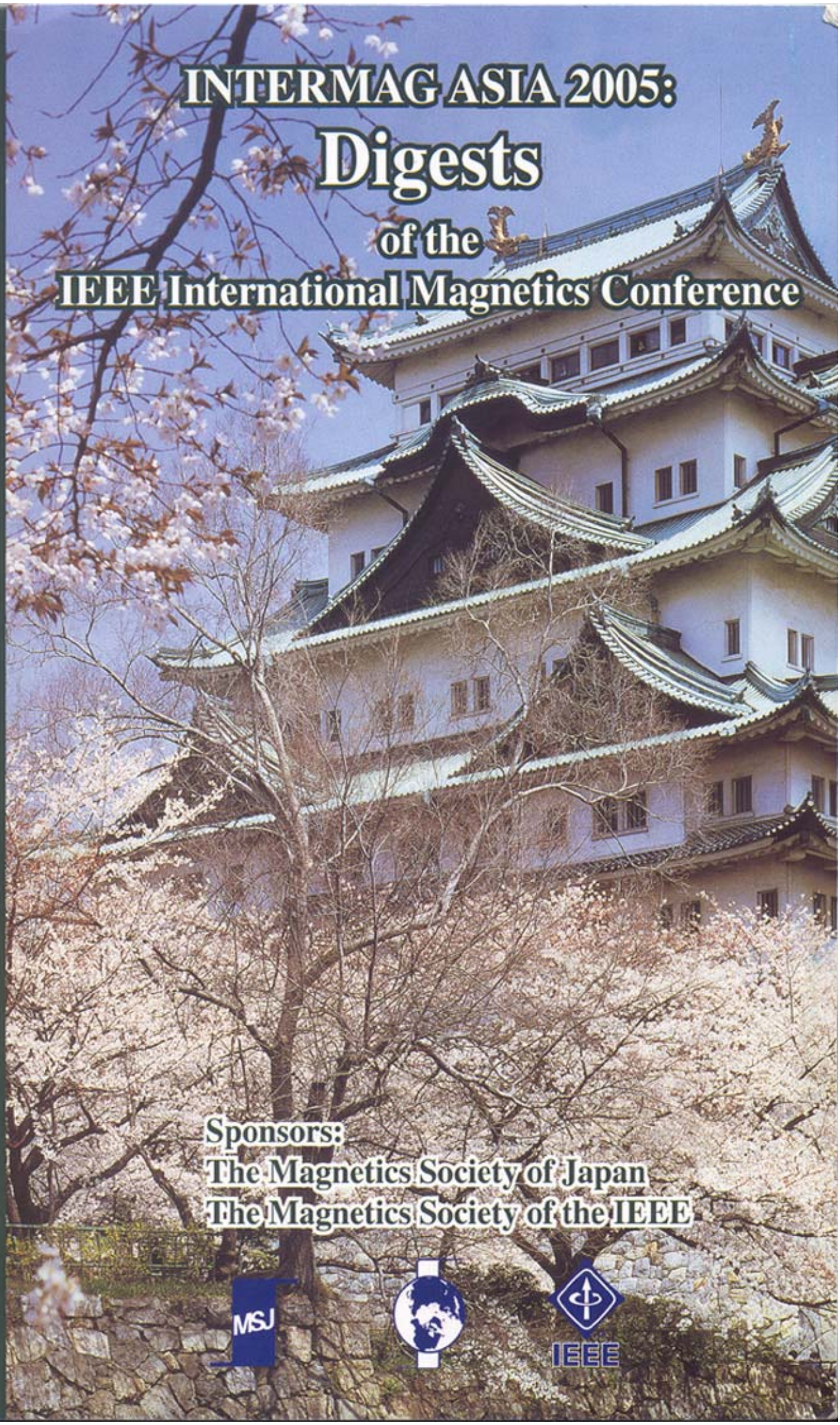
INTERMAG ASIA 2005:
Digests
of the
IEEE International Magnetics Conference

The 2005 IEEE International Magnetics Conference
April 4 – 8, 2005 • Nagoya Congress Center • Nagoya, Japan



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Anisotropic in-field Mössbauer spectra for $La_{1/3}Sr_{2/3}FeO_{2.96}$ above its charge disproportionation temperature

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The charge disproportionation (CD) was first observed by Mössbauer spectroscopy in the sample $La_{1/3}Sr_{2/3}FeO_3$, in which the Fe ions could take non-integral charge states between Fe^{3+} and Fe^{5+} depending on the composition [1]. Subsequently Battle *et al.* found a first-order like transition from a high-temperature paramagnetic average-valence state in which all Fe ions are electronically equivalent to a low-temperature antiferromagnetic mixed-valence state [2]. Park *et al.* reported a sudden change in the resistivity accompanied by the CD transition [3]. The charge state above the CD transition temperature can be explained by an itinerant nature of the σ^* band derived from the e_g electron in high-spin Fe^{4+} ($t_{2g}^3\ e_g^1$). An electron can hop from one Fe to another, generating both the intermediate valence state and the metallic conductivity. Battle already mentioned about the asymmetric line-shape of the paramagnetic singlet at 290K, which indicated that the obtained spectrum could consist of several components. Now, it will be of interest to observe the change in Mössbauer spectra above the CD transition temperature under external field. In this paper, we present results of external field Mössbauer spectroscopy for $La_{1/3}Sr_{2/3}FeO_{3-y}$ in order to see how the external magnetic field affects the CD state above its transition temperature.

The polycrystalline $La_{1/3}Sr_{2/3}FeO_{3-y}$ was prepared by the conventional solid-state reaction method. In order to prevent any inaccuracy in composition involved when using carbonate powder containing moisture, pure strontium-oxide powder was precisely weighed in a glove box filled with N_2 gas. The crystal structure and oxygen non-stoichiometry were checked by X-ray diffraction (XRD) and chemical analysis, respectively. A Mössbauer spectrometer of a conventional transmission-type was used in the constant acceleration mode. External magnetic field was applied both parallel and perpendicular to the direction of γ-ray propagation using a split magnet superconductor cryostat.

The following figures show Mössbauer spectra under 6 T for $La_{1/3}Sr_{2/3}FeO_{2.96}$ at 225 K, which is above the CD transition temperature. In the longitudinal geometry, a magnetic Zeeman component is superimposed to the central singlet. As the external magnetic field is

parallel to the γ-ray direction, the lines 2 and 5 are suppressed (the lines 3 and 4 are overwhelmed by the strong central singlet). It is clear that there is still a considerable fraction of the Fe nuclei that does not experience the applied field. This is an evidence of the fast electron-transfer among Fe ions. When such a fast hopping of electrons exists, fluctuations of hyperfine parameters on ^{57}Fe nucleus will take place, resulting in a relaxational singlet as can be seen in the spectrum. In the transverse spectrum, however, central singlet disappears and only magnetic component with intensity ratio 3:4:1:1:4:3 emerges. This means that magnetic moments are aligned perpendicular to the γ-ray direction. Absence of the central singlet in this case can be explained by the distribution of the γ-ray emission obtained on the basis of the stochastic calculation.

In conclusion, anisotropic Mössbauer spectra for $La_{1/3}Sr_{2/3}FeO_{2.96}$ above its CD transition temperature under external magnetic field were studied. Hopping of an electron between neighboring Fe ions occurs in such a way that the hyperfine magnetic field at the ^{57}Fe nucleus average out to zero. The observed magnetic component probably results from particular Fe ions in the neighbor of either an oxygen vacancy or a lattice imperfection that do not participate in the electron exchange process. And thermally generated hopping of delocalized carriers is not influenced by the application of magnetic field.

References

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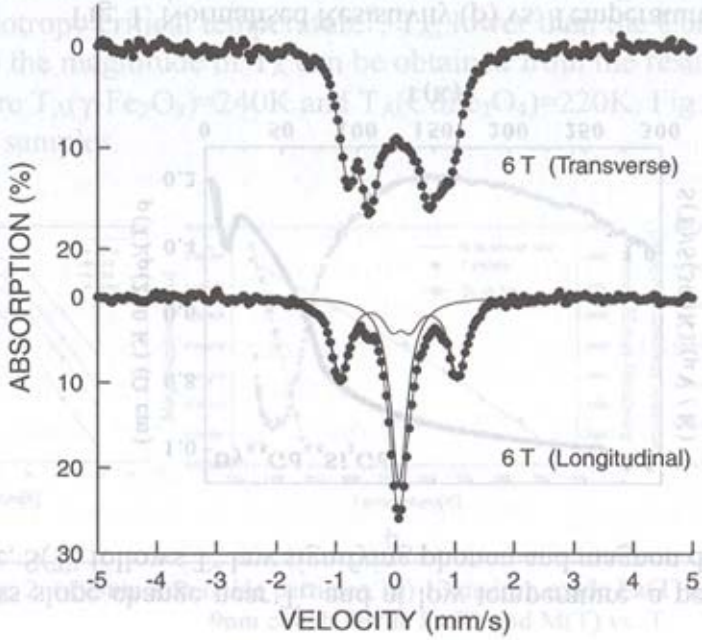

Fig. 1 In-field Mössbauer spectra under 6 T for $La_{1/3}Sr_{2/3}FeO_{2.96}$ at 225 K.