

## Charge ordering and Mössbauer studies of single crystal $R_{1/3}Sr_{2/3}FeO_3$ (R=Pr, Sm, and Nd)

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Single crystals of  $R_{1/3}Sr_{2/3}FeO_3$  (R=Pr, Nd, and Sm) were synthesized by the floating zone method and their magnetic properties and charge ordering (CO) transition related to lattice dynamics were systematically investigated. Mössbauer spectra of  $R_{1/3}Sr_{2/3}FeO_3$  were taken at various temperatures ranging from 12 K to room temperature. The charge disproportionation in  $Pr_{1/3}Sr_{2/3}FeO_3$  was detected below 190 K, in which two kinds of iron with valence states  $Fe^{3+}$  and  $Fe^{5+}$  were found with ratio of 2:1. The iron with valence state  $Fe^{4+}$  in  $Pr_{1/3}Sr_{2/3}FeO_3$  coexists at and above 150 K, and its ratio increased from 13% to 66% as the temperature rose. The  $(Nd_{1-y}Sm_y)_{1/3}Sr_{2/3}FeO_3$  ( $y = 0.0, 0.2, 0.4, 0.6, \text{ and } 0.8$ ) with least lattice distortion underwent a CO phase transition at and below  $T_{CO} = 163$  K and accompanying the charge disproportionation into nominally  $Fe^{3+}$  and  $Fe^{5+}$  sites as well as a canted antiferromagnetic spin ordering. In this charge ordering state, a sequence of  $Fe^{+3}Fe^{+3}Fe^{+5}Fe^{+3}Fe^{+3}Fe^{+5}$  exists aligned along the [111] direction of the pseudocubic perovskite structure. In this study, the CO at  $x=2/3$  disappeared in the case of R=Sm. Indeed, disappearance of the CO transition was detected by systematic decrease of a spontaneous magnetization with increase of  $y$  in the system of  $(Nd_{1-y}Sm_y)_{1/3}Sr_{2/3}FeO_3$ . This result shows that the charge ordering state was realized with strong hybridization between the Fe and O atoms.