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Relaxation effects and re-entrant spin glass behavior at low temperatures in natural strunzite, ferristrunzite, and ferrostrunzite

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Abstract

The temperature variations of the ferric and ferrous hyperfine fields in natural samples of strunzite, ferristrunzite, and ferrostrunzite are examined by Mössbauer spectroscopy between 4.2 K and their respective magnetic transition temperatures. The spectra of all three strunzite species are composed of two dominant sextets with equal contribution and with hyperfine fields at 4.2 K of approximately 48.5 and 48.0 T, respectively. They are assigned to Fe^{3+} cations in the Fe(1) and Fe(2) sites in the crystallographic structure. A third magnetic component, with field of 54.8 ± 0.1 T, is well resolved in the 4.2 K spectrum of ferristrunzite and is attributed to Fe^{3+} in the Mn sites of the structure. This third ferric contribution also appears in spectra of strunzite and ferrostrunzite, however with smaller relative spectral areas as compared to ferristrunzite, i.e., ~10% and ~3% of total spectral area, respectively, where it is 30% for ferristrunzite. For ferrostrunzite, two additional magnetic components are recognized. They are both attributed to Fe^{2+} substituting in the Mn sites. Their hyperfine fields at 4.2 K are 21.8 ± 0.5 T and 28.6 ± 0.5 T, respectively, and their relative contributions 11% and 15%. With increasing temperature, enhanced asymmetric broadening of the absorption lines is observed for all three samples. This phenomenon is due to relaxation of the magnetic moments, thus causing gradually broader distribution ranges of the respective hyperfine fields. The spectra have been fitted with superpositions of model-independent hyperfine-field distributions. The temperature dependencies of the maximum-probability hyperfine fields of the two ferric subspectra associated with the Fe(1) and Fe(2) sites are interpreted in terms of the molecular field theory taking into account the occurrence of exchange magnetostriction. The variations with temperature of the magnetic hyperfine fields acting at the Mn sites indicate that the magnetization of the Mn sublattice shows re-entrant spin glass behavior. This feature is most prominent for the strunzite sample.

Keywords: Strunzite, ferristunzite, ferrostrunzite, Mössbauer spectroscopy, hyperfine field