Magnetic nanoparticles of iron oxides, particularly those of ferric oxides, magnetite, substituted variants, including functional derivatives, have attracted significant interest because of promising capabilities in emerging diagnostic imaging methods and novel therapeutic interventions. In addition to the rapid development of syntheses and functionalization methods, a comprehensive understanding of their fundamental physical characteristics and the complex link among composition, microstructure, magnetic properties, and relaxation characteristics is pivotal for the rational design of well-defined magnetic nanoparticle systems. Generally, the magnetic behavior of nanoparticles deviates from bulk materials not only due to finite-size and surface effects but also by the occurrence of new crystal structures (polymorphs) and metastable states, such as non-equilibrium cation distribution. The doctoral thesis investigates the key physical characteristics of selected nanoparticle systems (simple ferric oxides of various crystal structure and morphology; Zn-, Co- and Mn-substituted magnetite-maghemite; coated by various hydrophilic and biologically inert surface layers), primarily by ^{57}Fe Mössbauer spectroscopy and supplemented by other available techniques. For that purpose, Mössbauer spectroscopy that employs the atomic nuclei as local probes to examine the hyperfine interactions with their electron shells is advanced for a specific use in nanoparticle research (in particular for magnetic particle suspensions in a frozen state). To evaluate the attributes of magnetic nanoparticle suspensions critical for applications in magnetic resonance imaging, magnetic particle imaging, magnetic hyperthermia, and targeted drug delivery, a magnetic particle spectrometer, an emerging method that exploits the nonlinear response of magnetic nanoparticle suspensions when subjected to an alternating magnetic field, was developed, with a broad spectrum of operating frequencies and magnitudes of the driving field.